Characterizing Traffic-Related Air Pollutant Dynamics in a Near-Road Environment

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Jennifer Lynn Moutinho

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Characterizing Traffic-Related Air Pollutant Dynamics in a Near-Road Environment

Approved by:

Dr. Armistead G. Russell, Advisor
Environmental Engineering
Georgia Institute of Technology

Dr. Rodney Weber
Earth and Atmospheric Sciences
Georgia Institute of Technology

Dr. James Mulholland
Environmental Engineering
Georgia Institute of Technology

Dr. Jeremy Sarnat
Public Health
Emory University

Dr. Stefanie Sarnat
Public Health
Emory University

Approved on: November 7, 2018

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SUMMARY

Highly trafficked roads are major emissions sources due to tailpipe and evaporative emissions, brake and tire wear, and resuspended road dust leading to high levels of localized pollution. While the locally high levels of pollutants are due mostly to primary emissions of carbon monoxide (CO), nitrogen oxides (NOx), and particulate matter ($PM_{2.5}$), tailpipe and evaporative emissions can also lead to the formation of secondary pollutants including nitrogen dioxide as well as particulate matter and ozone. A number of factors including meteorological conditions, traffic patterns, and road features drive the spatial and temporal dynamics of traffic-related air pollutants (TRAPs) occurring in the near-road environment. While elevated concentrations of TRAPs are still found in the near-road environment, better controls and fuel regulations have decreased mobile tailpipe emissions despite an increase in the number of vehicles on the roadways and an increase in fuel consumption. As tailpipe emissions become a smaller contributor to traffic-related air pollutants in the near-road environment, alternative methods need to be considered in order to characterize population exposure to traffic-related air pollutant concentrations.

This research examines the near-road environment to understand the impact of roadway emissions on adjacent microenvironments and the potential implications related to quantifying exposures for individuals. The objectives are to understand what factors drive the near-road traffic-related air pollutant concentrations, to characterize exposure to primary vehicle tailpipe emissions using alternative multipollutant metrics, and to model

mobile emissions to quantify exposure variability based on concentration changes and individuals' movements throughout the day. A 15-lane highway though Atlanta, Georgia is one of the busiest highways in the country with an annual average daily traffic (AADT) count of 330,000 vehicles. The highway runs north-south through downtown Atlanta and is a roughly 10-mile stretch where Interstate 75 and Interstate 85 merge (I-75/I-85). This stretch of highway also runs along the eastern border of the Georgia Institute of Technology (GIT) campus. Monitoring at six locations on the GIT campus within two kilometers of the highly trafficked interstate, including two near-road sites, characterized factors that drive the spatiotemporal variability of individual traffic-related air pollutants and two multipollutant metrics. The suitability of single pollutant concentrations and multipollutant indicators are evaluated as personal exposure metrics for small cohort epidemiological studies addressing the limitations of using a single near-road monitor to qualify exposure. This dissertation also evaluates the accuracy of atmospheric dispersion modelling at different temporal and spatial scales. Measured spatial gradients provide data to develop ways to calibrate the model results to improve the accuracy of simulated hourly exposure concentrations.

Observations found relatively low concentrations in the near-road environment with diurnal profiles driven more by meteorological factors than hourly traffic count. Measurement differences at two near-road monitoring locations along the same road segment highlight the importance of monitoring site placement emphasizing limitations to the use of any individual monitoring locations for generalizing the characterization of population exposure near urban roadways. As vehicle emissions continue to decrease, even in the near-road environment, mobile source emissions are not the dominant source of

pollutants such as $PM_{2.5}$, and the fraction of NOx and CO from mobile sources are declining. While regional sources have a growing contribution to pollutant concentrations in the near-road environment, a heterogeneous spatial distribution still exists near (within approximately two kilometers of) highly trafficked roadways making it difficult to characterize exposure to the single source.

Multipollutant indicators can characterize exposure to a single source using a statistical emissions-based metric or a metric of particulate matter toxicity measured at the central monitors. The statistical metric was a more stable method for assessing exposure to primary tailpipe emissions. The metric of toxicity based on oxidative potential did not appear to be a good measure of primary mobile tailpipe emissions since it was sensitive to secondary species formed from vehicle emissions precursors, as well as other sources such as biomass burning. These metrics were assessed for their ability to increase correlations between monitoring sites compared to correlations observed in single primary pollutants. A higher correlation between two monitoring sites suggests that the metric is more representative of exposure around and between the two sites. Alternatively, the spatial resolution of pollutant concentrations could increase to improve estimates of personal exposure through the increase in monitoring sites or the use of air quality models. Dispersion models can provide complete spatial fields for exposure assessments. R-LINE, a steady-state dispersion model, was applied to the Atlanta, GA region, using a detailed, link-based mobile source inventory for CO, NOx, and primary $PM_{2.5}$. However, in the application here, the model was biased high near the main highway when estimating hourly concentrations. Several difference methods were used to reduce the model bias for accurate hourly exposure assessments. Through this dissertation, a clearer understanding of the

near-road spatial distribution of primary tailpipe emissions developed a better understanding of microenvironments that exist along congested roadways.

This dissertation focuses on characterizing the temporal and spatial variability of air pollutants observed in the near-road environment, and the usefulness of various methods to characterize population exposure such as multipollutant metrics and dispersion modeling. Most of the analysis was conducted as part of the Dorm Room Inhalation of Vehicle Emissions (DRIVE) Study aimed at characterizing exposure to vehicle-related emissions. A key part of the study was characterizing the dynamics of TRAPs. Chapter 2 explains the larger study that utilized the measurements and analysis presented in this thesis. Chapter 3 describes the temporal trends observed in the near-road measurements and explores the differences observed between two near-road monitoring sites along the same road network highlighting how important physical site characteristics are to microenvironment concentrations. Chapter 4 focuses on the applicability of two multipollutant indicators to capture the temporal and spatial trends observed in the single pollutants. Measurement trends capture all local and region sources, therefore the metrics were also assessed for their ability to capture exposure to mobile sources focusing on how this may lead to differences in the trends when compared to the single pollutants. Chapter 5 addresses the use of dispersion models to link single source emissions to their contribution to local pollutant concentrations. This chapter also addresses the biases observed in the model output and simple correction methods that can be used to limit this bias. Finally, Chapter 6 provides a summary of the conclusions reached in each chapter of this dissertation and the possible directions future research could go to build off the work presented here.

CHAPTER 1

INTRODUCTION

1.1 Near-road Environment

In urban areas, highly trafficked roads are major emission sources that lead to high levels of traffic related air pollutants (TRAPs) throughout cities with localized levels near major arterials elevated above urban background concentrations (Beckerman et al., 2008). Such emissions can also interact chemically and physically with other pollutants pre-existing in the roadway environment. Exposure to individual TRAPs have been linked to adverse health effects (EPA, 2010c, 2011). Specifically, particulate matter has been associated with 7% of cardiorespiratory mortality and respiratory cancer combined according to the World Health Organization's Global Burden of Disease (WHO, 2002). Since motor vehicle emissions contribute about 16% to the particulate matter concentrations in ambient air, the overall association between motor vehicle emissions and mortality is likely to be substantial (HEI, 2009c). Adverse health effects, including the exacerbation of asthma, have been linked to primary vehicle tailpipe emission in populations exposed in the near-road environment (Brugge, Durant, & Rioux, 2007; EPA, 2001, 2009, 2010a, 2016; HEI, 2009c; WHO, 2005).

Elevated air pollutant concentrations exist within near-road microenvironments due to vehicle exhaust and evaporative emissions along with mechanically-generated emissions. While concentrations of nitrogen oxides (NOx) and carbon monoxide (CO) as

well as primary fine particulate matter are elevated near heavily trafficked roads, improved vehicle engine technologies, emissions control systems, and fuel regulations have led to reduced mobile source emissions (Ayala, Brauer, Mauderly, & Samet, 2012; Henneman, Holmes, Mulholland, & Russell, 2015; Karner, Eisinger, & Niemeier, 2010; Vijayaraghavan, DenBleyker, Ma, Lindhjem, & Yarwood, 2014; Zhu, Hinds, Kim, & Sioutas, 2002). Although annual national mobile source fuel consumption increased 18% between 1996 and 2006 with on-road vehicles accounting for 85% of the fuel consumption, NOx emissions decreased 49% from on-road gasoline vehicles and only increased 7% from on-road diesel vehicles (Dallmann & Harley, 2010). Across the urban southeast from 2000 to 2011, on-road mobile source NOy and CO contributions decreased about 50% while vehicle miles traveled (VMT) have increased by 18% (Blanchard, Tanenbaum, & Hidy, 2013; Vijayaraghavan et al., 2014). Reductions in mobile emissions have led to a 39% decrease in $PM_{2.5}$ mass at urban Georgia sites from 2002 to 2013 (X. X. Zhai, Mulholland, Russell, & Holmes, 2017). While vehicles remain a significant source contributing to near-road concentrations, the near-road environment is changing as a result of the decreased emissions (Batterman, 2013; Chow, Watson, Lowenthal, Chen, & Motallebi, 2011).

Within the United States, 58% of the population lives in urbanized areas with a population over 200,000 (GDOT, 2013). Further, approximately 19% of the US population lives near a high-volume road (Rowangould, 2013). Historically, the highest concentration of pollutants and therefore the highest risk of exposure occur near roads (Beckerman et al., 2008). Tail pipe emissions contain or lead to a range of primary and secondary pollutants including carbon monoxide, nitrogen dioxide, particulate matter, and ozone. Growing concern for the possibility of high concentrations of nitrogen dioxide (NO_2) in the near-

road environment motivated the U.S. Environmental Protection Agency (EPA) to require the implementation of a national near-road monitoring network (EPA, 2010c, 2011). The near-road monitoring network focuses on locating monitors within 50 meters of heavily trafficked roads in urban cores around the nation in order to improve exposure assessments to traffic emissions for populations spending time on or near these highways (Batterman, 2013; EPA, 2012).

The implementation of the network occurred in three phases based on core-based statistical area (CBSA) populations. The first phase required each urban center with a population above one million to establish a monitoring site for NO_2 before January 1, 2014. Phase two required an additional site to be established by 2015 in urban areas with a CBSA population greater than 2.5 million or in areas with a road segment that has an annual average daily traffic (AADT) greater than 250,000. Phase two also required the monitoring sites located in urban areas with a population greater than 2.5 million to measure continuous CO as well as $PM_{2.5}$ following integrated filter FRM or continuous FEM methods. The third phase required urban centers with a CBSA population between 0.5 million and one million to establish a site for measuring NO_2 concentration by 2017. These site have since been removed from the network. Phase three also required all sites in urban centers with a population between one million and 2.5 million to monitor CO and $PM_{2.5}$ concentration. In total phase one and two established 75 sites, of which 60% of the sites are within 20 meters of the target highways and 90% of the sites are within 30 meters of the target highways. Of the hourly NO_2 concentrations reported for monitoring sites with at least 75% data completeness by quarter, only five hours exceeded the hourly standard of

100ppb, potentially due to extreme weather events or unusually high idling traffic near the site (DeWinter, Brown, Seagram, Landsberg, & Eisinger, 2018).

With a population above 2,500,000 and a highway with an AADT above 250,000, Atlanta, GA established two monitoring locations. One site was placed along I-75/I-85 in central Atlanta and the other monitoring location is along I-285 about 15km southeast of downtown Atlanta. The central site was established as part of phase one; with some delay, NO_2 and CO concentration measurements began June 15, 2014. Located on the Georgia Institute of Technology campus, the site was located within a line of trees. The inlet was two meters from highway within the tree line and 4.5 meter above the highway. The second monitor was established as part of phase two and began monitoring NO_2 for January 1, 2015. The monitoring site is located 30 meters from the interstate in an open grass field.

Historically NO_2 concentrations have exceeded the hourly standard level in the near-road environment. Where high NO_2 measurements are still a concern, vehicle emissions are the major contributing source (Lin, Feng, & Heal, 2016). Elevated concentrations are effected by a number of factors including traffic volume, vehicle types, local meteorology conditions (Karner et al., 2010), and local topography including natural (R. Baldauf, 2017) and built highway features (R. Baldauf, 2017). These conditions can lead to a wide range in pollutant concentrations along different roadway segments within the same urban environment (McAdam, Steer, & Perrotta, 2011).

1.2 The Near-road as a Multipollutant Environment

Individuals are exposed to a mixture of air pollutants at different ratios depending on the local and regional sources. While individual pollutants have been linked to adverse

health effects, less is understood about the confounding effects of exposure to pollutant mixtures or which component in particulate matter has a greater relative toxicity. In addition to improving the scientific understanding of the combined effects of simultaneous exposure, methods need to be further developed to help set regulatory standards based on the human health risks of air pollution mixtures as a whole. While the development and application of multipollutant research may take time, the Environmental Protection Agency (EPA) has begun supporting this idea by revising the National Ambient Air Quality Standards (NAAQS) for four criteria pollutants in a more coordinated fashion through clustering the standard reviews. For past revisions, the EPA has addressed each pollutant individually allowing the states to take action to reduce the pollutants independently. Further, they separated out the welfare reviews of sulfur and nitrogen oxides. By revising the health-based standard levels for nitrogen dioxide, sulfur dioxide, ground-level ozone, and fine particles within a 16-month period, the EPA was building on the knowledge that pollutants often share the same source (Greenbaum & Shaikh, 2010). States, in turn, were encouraged to implement multipollutant reduction strategies at the same time to address such sources. The EPA has also had a long term regulatory interest in understanding and reducing near-road air pollution by passing the Mobile Source Air Toxics (MSAT) Rule recognizing locations near highly-trafficked roads as important areas for MSAT exposure (EPA, 2001).

In spite of the more recent movement to understand and address air pollution as a multipollutant mixture, the regulations are still formulated on a single pollutant basis. The US Clean Air Act and its Amendments laid out a regulatory structure that led to a focus on specific indicator pollutants and their known health effects. While it tends to be easier to

sample and link individual pollutants to adverse health effects, near-road pollution tends to be highly heterogeneous which has prompted studying TRAPs with a more multipollutant perspective (HEI, 2009c). Using a multipollutant framework provides opportunities to characterize exposure metrics for populations exposed to the mixture of TRAPs commonly found in the near-road environment (Greenbaum & Shaikh, 2010; Johns et al., 2012; Mauderly et al., 2010; Solomon et al., 2012; Vedal, 2011). An improvement to the understanding of the association between on-road emissions sources and numerous adverse health effects is particularly important as the near-road environment changes.

Epidemiological studies have used source-apportioned measures of primary mobile source emissions to estimate health risks associated to a mixture of pollutants (N. A. Janssen et al., 2011; Ostro, Feng, Broadwin, Green, & Lipsett, 2007; S. E. Sarnat et al., 2008). Alternatively, exposure studies can utilize metrics that combine concurrent measurements or assess biological response through measurements of oxidative stress. However, as the near-road environment changes, the spatial and temporal variability of single-pollutant and multipollutant metrics will also change effecting the accuracy of traditional methods.

1.3 Dispersion Modeling to Characterize Pollutant Dynamics in the Near-road Environment

With emissions decreasing from vehicles, tailpipe emissions are no longer the majority contributing source to the traffic-related air pollutant (TRAP) concentration measurements in the near road environment. Air quality dispersion models can provide simulated concentrations of primary species for single sources to help determine the

contribution of mobile emissions on ambient pollutant concentrations. While a variety of different computational frameworks have been used, a widely used approach is using models based on the Gaussian dispersion equation. The research line (R-LINE) source dispersion model was designed specifically for mobile, road-based emissions. The model generates hourly concentrations for each specified point location by assuming steady-state dispersion from a series of point sources representing the roadway emissions.

One limitation to Gaussian dispersion models, such as R-LINE, is the lack of chemical reactions or other species sinks. The inability for dispersion models to capture nonlinear chemical reactions leads to a limited set of primary species that can be modeled. This limits the models ability to capture the ozone cycle and its interaction with nitrogen oxides in the near-road environment. Chemical transport models help address this limitation, however have their own limitations such as requiring additional computing resources and proving grid-based concentrations at a courser resolution than dispersion models. In both cases, models are able to provide a complete spatial concentration field providing an improvement over limited monitoring sites and can assess the impact of individual sources providing direct source-receptor relationships, where sampling methods capture the total pollutant concentration for all sources. In order to assess exposure to the complex traffic network within an urban area, some level of modeling is necessary.

CHAPTER 2

THE DORM ROOM INHALATION TO VEHICLE EMISSIONS (DRIVE) STUDY

The DRIVE study was designed to measure traffic related air pollutants (TRAPs) and assess the use of multipollutant metrics to understand the exposure-to-dose pathway of TRAPs in the near-road environment. The Georgia Institute of Technology (GIT) campus provided a location to set up monitoring sites to evaluate the suitability of different metrics as surrogates for traffic exposure. The campus also facilitated the development of a small cohort for quantifying personal exposure for students living and studying on the campus.

The GIT campus is in the geographic core of Atlanta, GA with its eastern edge bordering a major highway known locally as the "Downtown Connector." The Downtown Connector is the name given to the stretch of two major interstate highways, I-85 and I-75, that converge for 10 miles through the center of Atlanta. This stretch of highway has 15 lanes and is a classic traffic emission hotspot with an average of 320,000 vehicles and about 16,000 trucks per day (GDOT, 2014). The Downtown Connector is an excellent near-road setting to better understand pollutant gradients (Yan et al., 2009). While numerous smaller roadways surround the campus, the Connector is the dominant mobile emissions source with an AADT count at least 15 times that of the other roads in the area. In addition, GIT has two groupings of student dormitories. One is in close proximity to the Downtown Connector, only 30m from the highway. Another group is removed from major roads and about 1.2 km from the highway. As such, this makes the campus an ideal location for

identifying and characterizing near-road pollutant dispersion and emission-to-exposure pathways.

2.1 Measurements

Intensive sampling occurred from September 2014 to January 2015 at four locations on the GIT campus as well as at two sampling sites that provided long-term data (Figure 2.1). Outdoor measurements were conducted at a near-road site located 5m from the highway (NR DRIVE), two dormitories, and a roof top lab. The dormitories collected both outdoor and indoor concentration measurements 30m from the highway (Near Dorm) and about 1.2 km west of the highway (Far Dorm). The roof top (RFT) sampling location was 500m from the highway in a fourth floor lab and provided measurements from reference instrumentation. Additionally, existing and ongoing monitoring sites were used to better assess the pollutant gradients: a Georgia Department of Natural Resources near-road monitoring site (NR GIT) and a site located 2.3 km west of the highway (UB).

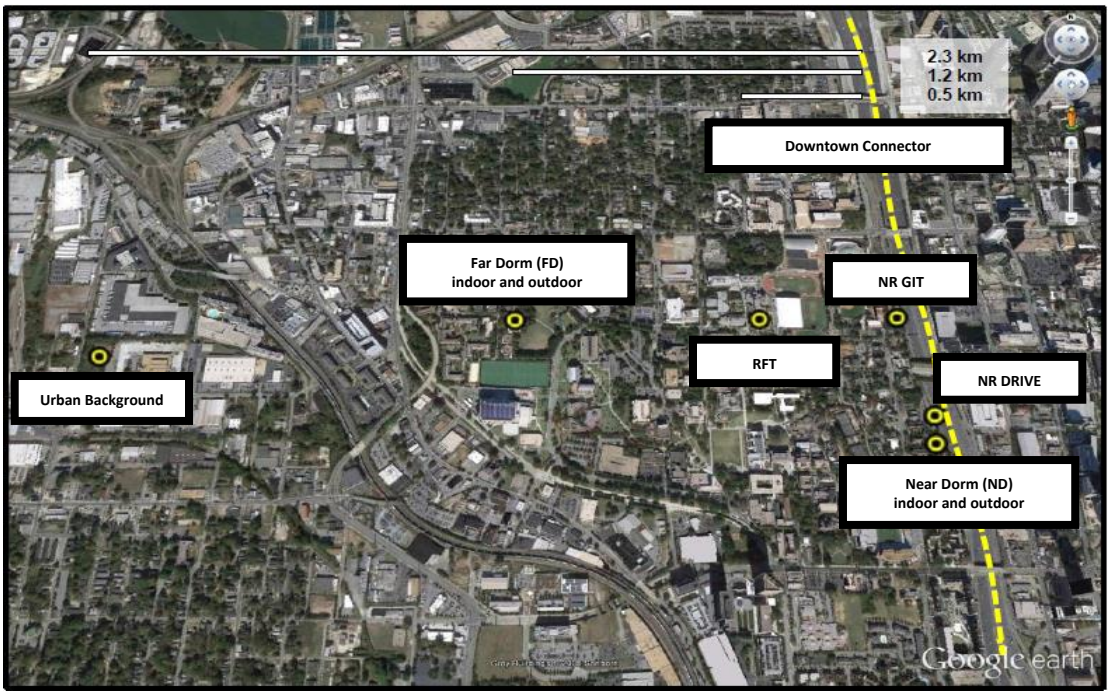

Figure 2.1 Map of Sampling Locations. NR GIT: Near-road Monitoring Network site on the GIT campus; NR DRIVE: Near-road DRIVE site; ND and FD: Near dorm and far dorm with ambient and indoor sampling; RFT: Roof-top sampling site; UB: Urban background site, Jefferson St SEARCH site

Pollutant measurements conducted included both commonly monitored TRAPs (CO and NOx), as well as primary species (EC) and secondary species related to traffic emissions (O_3) with the instrumentation varying between location (Table 2.1). Traditional primary traffic-related pollutants, including NOx, CO, and BC, were measured continuously at each sampling location. At the NR DRIVE site, additional instruments measured O_3 and direct NO_2. In addition to continuous and semi-continuous measurements, 48-hour quartz and Teflon filter-based measurements were collected for EC, OC, and $PM_{2.5}$. The filters were also used for oxidative potential to measure reactive oxygen species using an acellular dithiothreitol (DTT) assay from the water-soluble fraction of $PM_{2.5}$

measured. All species concentrations, meteorological parameters, and traffic data were measured and analyzed at local standard time (LST); however, daylight savings time did end on November 2, 2014 during the study monitoring period. A total of 55 instruments were utilized providing air pollutant concentration data at six measurement sites.

The NR DRIVE was located 5m from the closest highway lane and served as an anchor site for this study. Outdoor measurements were collected with an inlet 3m from the ground using a highly instrumented stationary trailer located in an open parking lot. The parking lot was level with the highway on the southbound side and there was no built or natural barrier around the site. On the northbound side of the highway a 5m wall runs along the east side since the local streets are at an elevation 5m above the highway. At the two residential buildings continuous sampling alternated indoor and outdoor air every 15-minutes using a three-way valve. The Near Dorm (ND) sampling was conducted out of a room in the basement of the Perry-Matheson dorm about 100m west from the Downtown Connector and 90m south of the stationary roadside site. A plexiglass insert placed in the window for the outdoor (NDO) inlets sampled from about 0.5m off the ground. The indoor (NDI) sampling inlet was about 0.25m off the rug flooring. Every 15-minutes a compressor changed the three-way valve positions so that the sampling would switch from indoor to outdoor. The Far Dorm (FD) was 1.2km from the highway and not located near any other major traffic emission sources. The sampling was in the Woodruff Dorm and was conducted out of a first-floor room facing Northside Dr. NW and Tech Parkway NW. A similar plexiglass insert placed in the window for the outdoor (FDO) inlets sampled from about 2m off the ground. The indoor (FDI) sampling inlet was about 0.25m off the tile

flooring. The same timer and three-way valve system was used to switch between the indoor and outdoor sampling inlets alternating every 15 minutes.

Table 2.1 Summary of measurements conducted at each monitoring location

Site	Measure	Instrument Model	Frequency (n=target sample #)
NR DRIVE	CO	Thermo 48i	Continuous
	NO-NO_2-NOx	Teledyne 200A	Continuous
	NO_2	Aerodyne CAPS	Continuous
	O_3	Thermo 49C	Continuous
	BC	Magee Scientific Aethalometer	Continuous
	$PM_{2.5}$ Sulfate	ACSM	Continuous
	$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr) - 2/wk
	$PM_{2.5}$ OC and EC	Sunset Labs TOR with IMPROVE	Integrated (48-hr) - 2/wk
	Traffic count and composition		Continuous
	Meteorology (temp, RH, wind)	HOBO U30	Continuous
NR GIT	Gases: CO, NO-NO_2-NOx, $PM_{2.5}$ BC, O_3		Continuous
ND & FD	CO	Teledyne 300E	Continuous
	NO-NO_2-NOx	Thermo 42C Low Source	Continuous
	NO_2	Ogawa badges	Integrated (48-hr) - 2/wk * 2 dorms
	$PM_{2.5}$ BC	microAeth AE51	Continuous
	$PM_{2.5}$ CPC	TSI 3785 (Near); TSI 3022A (Far)	Continuous
	$PM_{2.5}$ Mass	GRIMM	Continuous
	$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr) - 2/wk * 2 dorms
	$PM_{2.5}$ OC and EC	Sunset Labs TOR with IMPROVE	Integrated (48-hr) - 2/wk * 2 dorms
RFT	CO	Thermo 48C Trace Level	Continuous
	NO-NO_2-NOx	Thermo 42i Trace Level	Continuous
	$PM_{2.5}$ BC	Thermo MAAP 5012	Continuous
	$PM_{2.5}$ Mass	TEOM 1400a	Continuous
	$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr) – 2/wk
	$PM_{2.5}$ OC and EC	Sunset Labs TOR with IMPROVE	Integrated (48-hr) – 2/wk
	Meteorology (temp, RH, wind)	Davis Vantage Pro2	Continuous
UB	Gases: CO, NO-NO_2-NOx, NO_2, O_3		Continuous
	$PM_{2.5}$ (mass, ions, OC, EC)		Continuous
	$PM_{2.5}$ (metals)		Integrated (24-hr) - 1-in-3 day
	Meteorology (temp, RH, wind)		Continuous
Students	NO_2	Ogawa badges	Integrated (48-hr) - 2/wk/subject * 6 subjects/wk
	$PM_{2.5}$ Mass	µPEM Nephelometer	Continuous
	$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr) - 2/wk/subject * 6 subjects/wk
	$PM_{2.5}$ OC and EC	Sunset Labs TOR with IMPROVE	Integrated (48-hr) - 2/wk/subject * 6 subjects/wk
	Location Tracking	GPS	Continuous
	Time-activity diary		4/wk/subject * 6 subjects/wk (n=288)
	Recall survey		1/wk/subject * 60 subjects/wk (n=720)
	Saliva		1/wk/subject * 60 subjects/wk (n=720)
	Bloods (plasma)		1/month/subject * 60 subjects/month (n=240)

A roof top lab in the Ford Environmental Science and Technology building serves as a reference continuous monitoring (RFT) site located in the middle of campus about 500m from the highway. The Georgia Department of Natural Resources (EPD) began operating a near-road monitoring site (NR GIT) as part of the EPA near-road monitoring network in June 2014. The NR GIT site is located about 70m north of the stationary roadside site (NR DRIVE) along the Downtown Connector. While the site sampling inlet is located 5m west of the highway and 3m from the ground, similar to the roadside site, the state sampling site is located within a line of trees leading to vegetation impacts on the measured concentrations. Finally, the furthest site from the highway (UB) is the Jefferson St. site operated by Southeastern Aerosol Research and Characterization (SEARCH) and is generally considered representative of Atlanta urban background pollutant concentrations (Edgerton et al., 2005; Liu, Wang, Russell, & Edgerton, 2005; Solomon et al., 2003).

Since the CO concentration measured is affected by changes in ambient temperature and the sampling rooms were not held as a constant temperature throughout the day, the instrumentation needed to measure zero air every 15-minutes. The sample cell and detector must be maintained at a constant temperature in order for the detector to keep a stable background. Fluctuations of more than 1°C can cause the volume of the air to change leading to a drift in the baseline giving false readings at low levels. The actual CO concentration was calculated by taking the difference between the ambient CO measurement and the zero air CO measurement to account for the effects of temperature change that resulted in a diurnal profile in the baseline. The CO instrumentation at the NR DRIVE, ND, FD, and RFT sites were modified by adding a CO scrubber to provide zero

air to the instrumentation (Parrish, Holloway, & Fehsenfeld, 1994). The scrubber consists of a catalytic converter that uses 0.5% palladium on alumina spheres heated to 180°C to convert all CO to CO2 ($2\ CO + O_2 \rightarrow 2\ CO_2$). This scrubs all the CO from the ambient air sample stream creating a zero air stream. The recorded concentration is then the baseline value for the instrument. The NR GIT and UB sites use a zero air cylinder to provide a baseline reading.

2.2 Field Data Analysis and Verification

All field instrumentation used to measure continuous pollutant concentrations were evaluated and calibrated prior to field sampling. In order to compare concurrent pollutant measurements across the multiple sampling sites and ensure accurate concentrations during the sampling period, instruments measuring the same pollutant parameters were collocated both before and after the sampling period and consistently calibrated throughout the 13-week intensive field sampling period. In addition to verifying daily that the instrument was operating correctly, it was necessary to calibrate the instrumentation regularly throughout the sampling period about every two weeks. Calibration was done by varying the blend of pollutant gas from a cylinder of known concentration with a cylinder of zero air at given flow rates (Bios DryCal). Instrument collocations were conducted for continuous NO-NO_2-NOx and CO as well as integrated $PM_{2.5}$ mass and reflectance over a multi-day period, both before and after field sampling to assess method precision and potential instrument offset. Final concentration data reported were adjusted based on the time-weighted average of the calibration curves and the collocated measurements. Temporal corrections were also necessary for all the measurements conducted at the two dormitory sites due to a slight drift

(approximately 10-15 seconds per day) in the timer used to control the value that regulated the change in indoor and outdoor sampling.

Instrumentation at the NR GIT was maintained by the employees responsible for the other state required monitoring sites at the Georgia Department of Natural Resources and the UB site was maintained by Atmospheric Research and Analysis. This instrumentation is calibrated regularly based on the protocols required by the US EPA.

2.3 Personal Exposure Assessments

Panel-based and small cohort designs have proven to be especially effective approaches of investigating traffic related pollution and adverse health, given the ability to accurately measure both exposure and health parameters on an individual-level (Delfino et al., 2006; Delfino et al., 2008; McCreanor et al., 2007; S. E. Sarnat et al., 2012). Throughout the sampling period, researchers from Rollins School of Public Health at Emory University conducted personal exposure monitoring for two student cohorts, one group from each dormitory. Each week approximately six students from each cohort participated in personal exposure monitoring by collecting two 48-hour integrated samples for $PM_{2.5}$, EC, and NO_2. The personal sampling packs weighed approximately three pounds and were easily attachable to the strap of a backpack or bag to correspond to the breathing zone of subjects with minimal discomfort or alteration to their daily activity. Time-activity pattern data was also collected through portable global positioning system (GPS) trackers that were attached to the side of the pouch. GPS monitoring recorded participant locations continuously over the two consecutive 48-hour cycles. Locations or standpoints, which were defined as a point where the subject stayed for more than five minutes, were collected

in longitudinal and latitudinal points. Personal exposure monitoring data was used to better understand exposure level from traffic pollution due to personal outdoor exposure and infiltration into residential buildings. The logged GPS data was utilized to aid in quantifying time spent in various distances from the highway and time spent outdoors versus indoors.

2.4 Multipollutant Source Impact Metrics

The Integrated Mobile Source Indicator (IMSI) uses elemental carbon, carbon monoxide, and nitrogen oxides concentration measurements along with the fraction of these species emitted by gasoline and diesel vehicles to construct integrated indicators of gasoline and diesel vehicle impacts at a given location (Pachon et al., 2012). The total ISMI equation is:

$$ISMI = \frac{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{emis}\left(\frac{EC}{\sigma_{EC}}\right) + \left(\frac{NOx_{mob}}{NOx_{tot}}\right)_{emis}\left(\frac{NOx}{\sigma_{NOx}}\right) + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{emis}\left(\frac{CO}{\sigma_{CO}}\right)}{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{emis} + \left(\frac{NOx_{mob}}{NOx_{tot}}\right)_{emis} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{emis}}$$

where the scaled concentrations are normalized by the standard deviation (σ) of the pollutant concentration. The emissions estimate ratio of mobile source to total were developed using the Sparse Matrix Operator Kernel Emissions (SMOKE) Modelling System and are from the 4 km grid cell that includes the campus and the JST sampling site. A range of temporal integrations depending on 1-hour average, 1-hour maximum, and 24-hour average data will be developed for the ISMI time-series. The 1-hour average was used to compare the multipollutant indicator to the hourly single-pollutant concentration data.

The 1-hour maximum develops the metrics for assessing peak concentration and the 24-hour average will create comparable time-series to previous work.

The Fine Particulate Matter Oxidative Potential (FPMOP) multipollutant indicator represents a biologically-relevant cumulative method for characterizing pollutant exposures through its ability to produce reactive oxygen species (Ghio, Carraway, & Madden, 2012; N. Li, Hao, Phalen, Hinds, & Nel, 2003; Squadrito, Cueto, Dellinger, & Pryor, 2001; Tao, Gonzalez-Flecha, & Kobzik, 2003). An acellular assay measures oxidative activity of water-soluble particle components based on their ability to catalyze the transfer of electrons from dithiothreitol (DTT) to oxygen, generating superoxide radical anions. This assay mimics a similar process *in vivo* with physiological antioxidants in place of DTT. The rate of the reaction is proportional to the concentration of redox-active species in the collected particulate matter and is monitored by measuring the depletion of DTT. Components of aerosols, particularly carbonaceous compounds predominately from mobile emissions, may drive most of the fine particle matter oxidative potential and therefore toxicity. Prior studies have found an association between FPMOP and light-duty gasoline vehicles (Bates et al., 2015; Verma et al., 2014) and laboratory studies have found that engine emitted particles are DTT-active (McWhinney, Badali, Liggio, Li, & Abbatt, 2013; McWhinney, Gao, Zhou, & Abbatt, 2011).

Aerosol oxidative potential is considered more relevant to identifying the adverse health effects of particulate matter than mass concentration (Ayres et al., 2008). A variety of PM chemical constituents emitted by vehicles, such as black carbon, polycyclic aromatic hydrocarbons (PAHs), oxygenated PAHs such as quinones and hydroxyquinones, and transition metal species, have been linked to ROS generation in previous studies (Cheung

et al., 2010; de Kok et al., 2005; Surawski et al., 2010). The teflon 48-hour integrated filter samples collected at the four main sampling locations were used to measure the FPMOP indicator. The measured OP was normalized by volume of sampled air (expressed in units of nmol/min/m^3) to provide a measure of atmospheric concentration of aerosol OP. Alternatively, OP can be normalized by fine particulate mass to provide a measure of the intrinsic OP of the overall $PM_{2.5}$ (i.e., an indication of the "toxicity" of the $PM_{2.5}$, in terms of OP). Intrinsic OP also provides insight on contributions of specific sources to the overall aerosol oxidative potential.

2.5 Atmospheric Modeling of Mobile Source Impacts

The Research Line Source Model (R-LINE), an atmospheric line-source dispersion model, assessed the impact of emissions from the Downtown Connector on campus air quality through the development of pollutant gradients (Batterman, Ganguly, et al., 2014; Snyder et al., 2013). The model produced hourly CO, NOx, and $PM_{2.5}$ concentration fields at a 25m x 25m grid resolution from on-road mobile sources. It is important to note that there are limitations to the use of R-LINE and other dispersion models in this or other applications. The model simulates the concentration fields of primary pollutants as they are impacted from on-road emission; lacking the chemistry within the model means it does not include secondary $PM_{2.5}$ formation and ozone chemistry. Secondary $PM_{2.5}$ formation occurs photochemically as mobile VOC and NOx emissions react to form lower vapor pressure products (Gordon et al., 2013; May et al., 2014; May et al., 2013; Ranjan, Presto, May, & Robinson, 2012; Stelson & Seinfeld, 1982; Tkacik et al., 2014; Zhao et al., 2015, 2016) and ozone chemistry effects the concentration of NOx, as well as the fraction of NOx

that is converted from NO to NO_2. In addition deposition is not included as a factor that effects concentration, though over the short time scales involved here, that process is likely of lesser importance.

The emissions input included hourly traffic count from ARC data and local hourly meteorology. The R-LINE model domain included on-road mobile source emissions from the entire Atlanta region with an output fine resolution region that is a 6.5 km by 2.5 km area centered on the GIT campus. Emissions from roads outside of the fine grid domain area are included in the modeling, though the concentration maps generated focus on the fine grid area. Emission inputs used were the 2010 average, link-based, on-road mobile source emissions in the 20-county region surrounding metro Atlanta developed by the Atlanta Regional Commission (ARC) using their traffic demand and mobile source emissions modeling (D'Onofrio, 2015; X. Zhai et al., 2016). ARC estimated the emissions, in g/m/s, of carbon monoxide, nitric oxide, and fine particulate matter for 43,712 links based on modeled traffic volume, vehicle speed, and fleet demographics. MOVES (Ayala et al., 2012; Batterman, Burke, et al., 2014; EPA, 2010b, 2014) was then used to estimate annual emissions from each link.

The hourly meteorological input data was developed with AERMET (Cimorelli et al., 2005; EPA, 2004). The surface meteorological data measurements were from the National Weather Service at the Hartsfield-Jackson Atlanta International Airport (ATL) and preprocessed using AERMINUTE (EPA, 2015). The upper air data measurements were from the Peachtree City Falcon Field Airport (FFC). Since steady-state dispersion models tend to overestimate concentrations during calm air conditions, when the wind speed is less than 1 m/s, the EPA suggests resetting any wind speed less than 1 m/s to 1

m/s (EPA, 2000). Between September 1, 2014 and December 31, 2014, there were 88 hours with a wind speed below 1 m/s.

Additional corrections were necessary since ARC modeling was for 2010. Therefore, the 2010 link emissions were scaled to 2014 using the mobile emissions ratio of 2014 to 2010 from the Motor Vehicle Emission Simulator 2014 (EPA, 2014). The average diurnal emissions profile was also used to provide hourly link-based emissions. Accounting for the diurnal variation in emissions is important because in the evening when the boundary layer is low, if the annual average traffic volume is used, the estimated pollutant concentrations are biased high (X. Zhai et al., 2016).

CHAPTER 3

NEAR-ROAD VEHICLE EMISSIONS AIR QUALITY MONITORING FOR EXPOSURE MODELING

Exposure to vehicular emissions is linked to several adverse health effects. In response to the rising concerns, near-road monitoring is conducted to better characterize the impact of mobile source emissions on air quality and exposure in the near-road environment. As part of a study in 2014 to further understand exposure of individuals indoors and outside in the near road environment, an intensive measurement campaign collected traffic-related air pollutants (TRAPs) and related data (e.g., meteorology, traffic, regional air pollutant levels) in Atlanta along one of the busiest highway corridors in the US. The broader study included modeling of pollutant emissions across the metropolitan area, as well as quantifying personal exposures linked to human responses. The study aimed to better understand how well measurements from a near-road monitor relate to concentrations within the near-road environment around that monitor, and the resulting potential exposures, recognizing that the near-road environment is complex and pollutant gradients can be large. Given the complexity of the near-road environment, the study also aimed to characterize how closely two near-road monitors track each other and how this might influence the siting of other near-road monitoring stations. TRAP measurements, including carbon monoxide (CO) and nitrogen dioxide (NO_2) are analyzed at three roadside monitors in Atlanta, GA during 2014 and 2015. Two of the near-road monitors were

located within 300m of each other and a third was along a different, but still highly trafficked, highway about 15km southeast. While both meteorological and traffic conditions were monitored to assess the temporal impact of these factors on traffic-related pollutant concentrations, the meteorological factors drove the diurnal variability of primary pollutant concentration more than traffic count. In spite of their proximity, while the carbon monoxide and nitrogen oxide concentrations were correlated with similar diurnal variations, pollutant concentrations at the two closely sited monitors differed, likely due to the differences in the siting characteristics reducing the dispersion of the primary emissions out of the near-road environment. Overall, the near-road TRAP concentrations at all sites were not as elevated as seen in prior studies, supporting that decreased vehicle emissions have led to significant reductions, even along major interstates. Further, the differences in the observed levels show that use of single near-road observations will not capture pollutant levels representative of the local near-road environment and that additional approaches (e.g., air quality models) are needed to characterize exposures.

3.1 Introduction

Elevated air pollutant concentrations exist within near-road microenvironments due to vehicle exhaust and mechanically-generated emissions (R. W. Baldauf et al., 2013; Karner et al., 2010). Concentrations of traffic-related air pollutants (TRAPs), including nitrogen oxides (NOx), volatile organic compounds (VOCs), carbon monoxide (CO), primary fine particulate matter ($PM_{2.5}$), elemental carbon (EC), and organic carbon (OC), in particular have been found to be elevated near heavily trafficked roads (Richard Baldauf et al., 2012; Beckerman et al., 2008; Boogaard et al., 2011). Such emissions can interact

chemically and physically with each other and other pollutants pre-existing in the roadway environment, leading to a complex, multicomponent mixture (Saha, Reece, & Grieshop, 2018). While the near-road remains a potential high exposure environment, improved vehicle engine technologies and associated emissions control systems, and fuel regulations have led to reduced mobile source emissions affecting near-road concentrations (Ayala et al., 2012; Henneman et al., 2015; Karner et al., 2010; McDonald, Goldstein, & Harley, 2015; Vijayaraghavan et al., 2014). A decrease in mobile emissions in Georgia over the last decade has contributed to an estimated 30% reduction in PM from mobile sources (X. X. Zhai et al., 2017). While vehicles remain a significant source contributing to near-road concentrations, the near-road environment is changing as a result of the decreased emissions (R. W. Baldauf et al., 2013; Batterman, 2013; Beckerman et al., 2008; Blanchard, Tanenbaum, et al., 2013; Boogaard et al., 2011; Chow et al., 2011).

Exposure of populations to TRAPs has been extensively studied in near-road environments (Brugge et al., 2007; HEI, 2009a, 2009c; WHO, 2005). A growing concern for the possibility of high concentrations of nitrogen dioxide (NO_2), one of the National Ambient Air Quality Standard (NAAQS) pollutants, initiated the U.S. Environmental Protection Agency (EPA) to implement a national near-road monitoring network to specifically measure TRAPs (EPA, 2010c, 2011). The near-road monitoring network focuses on locating monitors near the most heavily trafficked roads in urban cores around the nation. One objective for the network was to improve exposure assessments to primary traffic emission for urban populations vulnerable to this pollution source. The implementation of the monitors began January 1, 2014 and as of January 1, 2015, there were 61 active monitoring sites, two of which were located within 10 m of two different

heavily trafficked highways in Atlanta, GA. These sites measure pollutants traditionally attributed to vehicle-related emissions and can be used to characterize elevated exposures in the near-road environment.

Historically NO_2 concentrations exceeded the hourly NAAQS in the near-road environment (Vijayaraghavan et al., 2014), though emissions controls are leading to reduced levels and exceedances (DeWinter et al., 2018). Where high NO_2 measurements are still a concern, vehicle emissions are the major contributing source (Lin et al., 2016). Elevated concentrations are affected by a number of factors including traffic volume, vehicle types, local meteorological conditions (Karner et al., 2010), and local topography including natural (R. Baldauf, 2017) and built highway features (R. W. Baldauf et al., 2016). These conditions can lead to a wide range in pollutant concentrations along different roadway segments within the same urban environment (McAdam et al., 2011).

The Dorm Room Inhalation to Vehicle Exhaust (DRIVE) study was conducted in Atlanta, GA in 2014 along one of the busiest highway corridors in the US to characterize factors leading to human exposures in the near-road environment, both indoors and outside, and to assess integrated and biologically-relevant traffic exposure metrics for applications in epidemiological studies (D. Liang et al., 2018; D. H. Liang et al., 2018). Utilizing measurements from the DRIVE study, this paper aims to characterize pollutant dynamics in the near-road environment and to examine how well one (or two in large metropolitan areas) near-road network monitors can characterize TRAP concentrations in complex, dynamic urban environments. This issue is addressed by focusing on the dynamics of TRAP observations in relationship to meteorology, traffic characteristics, and regional air pollution mixtures using two near-road monitoring sites in close proximity to each other

along the same highway segment located near downtown Atlanta. One site was part of the DRIVE exposure study, and the other site was part of the EPA Near-road Monitoring Network. The issue is further addressed by assessing factors driving TRAP concentrations between two EPA Near-road Monitoring Network sites within urban Atlanta along two different major highways. In particular, we compare observations of TRAPs from the two sets of near-road sites and quantify the impact of major factors on the near-road pollutant concentrations. Understanding how well a single monitor represents the concentrations across an urban area has importance in both regulatory as well as health assessment frameworks. Therefore, as part of this analysis we consider how the characterization of exposure and potential NAAQS exceedances might be different at two near-road monitors along the same road segment as well as two near-road monitors placed in different areas of an urban area.

3.2. Methods

Primary tailpipe emissions include carbon monoxide (CO), nitrogen oxides (NOx), and particulate black carbon (BC). Concentrations of these pollutants are used as tracers for the impact of on-road mobile emissions to urban environments. Measurements were conducted continuously along a major highway in Atlanta, GA at two near-road highway monitors located in relatively close proximity (about 300 m). One of the near-road locations was part of the DRIVE study and the second was part of the EPA Near-road Monitoring Network operated by the Georgia Environmental Protection Division (GA EPD) of the Department of Natural Resources since June 2014. This analysis first assesses hourly concentration measurements from two near-road monitoring locations along the

same highway segment and characterizes the major meteorological and traffic contributing factors. There were 54 cities in the EPA Near-road Monitoring Network in 2015 and six cities, including Atlanta, had two monitoring sites. While the second near-road site was not operating during the DRIVE study measurements, this site is included for context and comparison with the first EPA near-road site. Through comparing concentrations at the two sites within 300m of each other and the two sites along different major highways, the analysis can further assess how representative the EPA near-road network monitors are for understanding exposure to populations within the near-road environment of an urban area.

In addition to the near-road monitoring location, the DRIVE study included three other monitoring locations 5m, 1.5km, and 2.3km west of the highway source to capture the spatial gradient within the study domain. The spatial variability was further assessed temporally to show how the correlation between the near-road monitoring location and the other monitoring locations varied spatially and diurnally (D. H. Liang et al., 2018). Findings from the DRIVE study were consistent with other recent studies that have shown a decrease in near-road TRAP concentrations leading to an overall lower impact of major highways on local air quality (R. W. Baldauf et al., 2016; Kenagy, Lin, Wu, & Heal, 2016; X. B. Li et al., 2016; Richmond-Bryant et al., 2017; Smith et al., 2015). A more comprehensive discussion of the study can be found elsewhere (J. A. Sarnat et al., 2017), including a list of all pollutants measured.

3.2.1 Site descriptions

This study focuses on emissions and air quality along a segment of arterial interstate where Interstate 75 and Interstate 85 (I-75/I-85) have merged in the center of Atlanta,

Georgia (Figure 3.1). In 2014, this highway segment along which the two monitoring locations are situated had an average annual daily traffic (AADT) of 330,000, composed primarily of light-duty gasoline passenger cars and trucks. Heavy-duty diesel trucks made up approximately four percent of the average daily vehicles on this portion of the highway (GA DOT 2012). Surface streets to the east of the highway follow a gridded pattern with an average block length of 450 feet and an AADT of more than 15 times less than the AADT for the highway segment. The land west of the highway segment for 1.5 km is the Georgia Institute of Technology (GIT) campus with limited vehicle access and much lower AADT still. Within the study domain area, the Southeastern Aerosol Research and Characterization (SEARCH) network has maintained an urban background (UB) monitoring site since 1998 (Hansen et al., 2003). The site is located 2.3km west of the highway and is a long-term dataset that represents the historical Atlanta background concentration.

The near-road DRIVE sampling location (NR DRIVE) was located about a meter from the west side of the fifteen-lane highway (eight southbound and seven northbound) to the south of 10th Street and to the north of North Avenue (Figure 3.1). The monitoring site was located in a parking lot with less than 85 passenger vehicle spots and the vertical height from the highway to the parking lot was 0.5 meters. The nearby EPA near-road monitoring network site was located on the Georgia Institute of Technology campus (NR GIT) about 300 meters north of the NR DRIVE site location and about a meter from the west side of the highway. Trees were removed from the vegetation barrier to provide space for the site and a small, gated parking lot for about 100 passenger vehicles is to the west of the site. The second near-road monitoring network site (NR SDK) was located 30m from

the interstate circling Atlanta (I-285), about 14 km southeast of the city center and the NR GIT site as well as 2 km northeast of the GA EPD South Dekalb (SDK) state site. In 2015, this highway segment had an AADT of 144,000 with about 13 percent heavy-duty diesel trucks (GDOT, 2014). Truck traffic is required to use this route around the city leading to a higher concentration of diesel vehicles (13.1%) compared to the distribution observed on the highway segment measured by the near-road sites (4.2%).

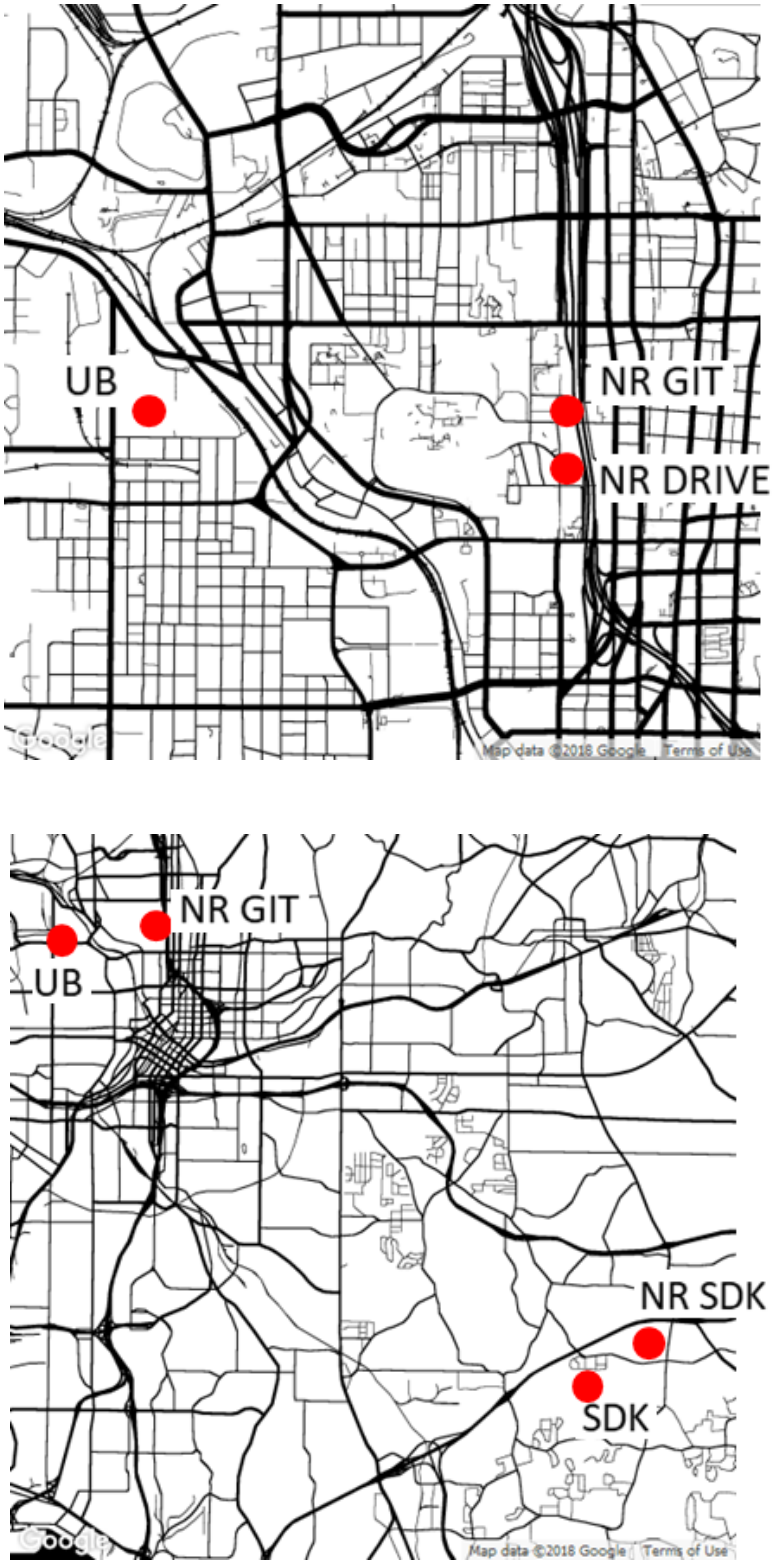

Figure 3.1 Map of sampling area. NR GIT site part of the EPA Near-road Monitoring Network in metro Atlanta, UB site part of the SEARCH Network, NR SDK site part of the EPA Near-road Monitoring Network in Atlanta, SDK site part of the GA EPD state monitoring network

3.2.2 Air quality instrumentation, meteorological characterization, and traffic data

Air quality monitors at the NR DRIVE location collected continuous ambient air samples from September 8, 2014 to January 5, 2015. All species concentrations, meteorological parameters, and traffic data were measured at local standard time (LST); however, daylight savings time did end on November 2, 2014. Continuous measurements of black carbon (Magee Aethalometer AE31), carbon monoxide (Thermo Model 48i), ozone (Thermo Model 49C), nitrogen oxides (Teledyne API 200A), and direct nitrogen dioxide (Aerodyne CAPS) provided concentration data for pollutants commonly associated with vehicle emissions. Real-time gas analyzers collected measurements at 5-second averaging periods and the real-time black carbon monitor collected at 2-minute intervals. Data was collected using DAQFactory and WinWedge Pro software. Multipoint calibrations, zero air, or span checks provided an assessment for accuracy throughout the study and were used in time-weighted adjustments to the data. The sampling inlet height was approximately 3m and was 7m from the closest highway lane. All continuous data were averaged to hourly levels to assess temporal variability differences between pollutants and possible indicators. Details of the instrumentation and quality assurance can be found elsewhere (J. A. Sarnat et al., 2017).

Continuous CO and NOx data from the NR GIT site began on July 1, 2014. The sampling inlet height was approximately 3m and was 6m from the closest lane. The hourly concentration data were downloaded from the EPA air quality system (AQS). The second EPA near-road monitor (NR SDK) had an operation start date of January 1, 2015. Hourly concentration data for BC and NOx from the NR SDK site and hourly CO and NOx data

from the SDK site were downloaded from the EPA AQS to assess the near-road sites in 2015.

Traffic vehicle count and speed data were obtained from the Georgia Department of Transportation Office of Transportation Data (GDOT, 2014). The vehicle count data were collected at a location on I-75/I-85 1.5 miles south of the measurement location using Automatic Traffic Records. No major on or off ramps are located between the traffic count location and the near-road monitoring locations.

Meteorological data collected at the NR DRIVE site (HOBO U30, Onset Corp) included wind speed, wind direction, temperature, and relative humidity. Wind speed and direction measurements used a cup anemometer and wind vane sensor. The wind rose (Figure 3.2a) from the NR DRIVE location found winds from the east (between 45 to 135 degrees) 77% of the time during the study period. With the NR DRIVE and NR GIT sampling locations west of the highway, winds from the east lead to downwind concentrations measurements. Additional wind measurements were collected at the NR GIT site (Figure 3.2b) and at the SDK site (Figure 3.2c) from January 1, 2015 to December 31, 2015. Mixing height observations were not available in the near-road environment, but can be a critical factor for pollutant dispersion. For this reason, mixing height data was modeled using the Weather Research and Forecast (WRF) model (NCAR) for the 4km grid that included the study domain.

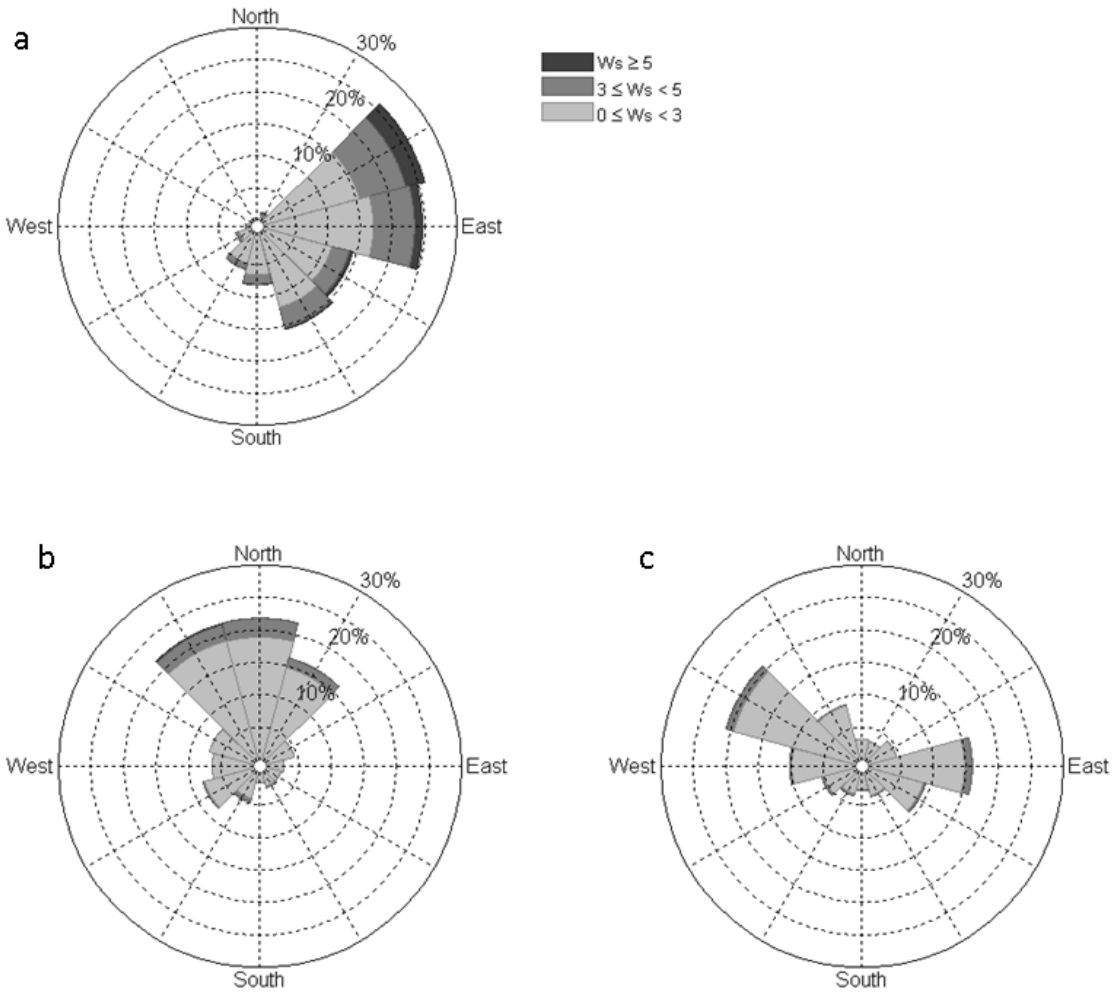

Figure 3.2 Wind rose of hourly observations at the (a) NR DRIVE site from September 8, 2014 to January 5, 2015 (b) NR GIT site from January 1, 2015 to December 31, 2015 (c) SDK site from January 1, 2015 to December 31, 2015

3.2.3.Multivariate regression modeling

To assess the factors that affected the temporal variability in the concentration of each TRAP, this study used a multivariate, linear, mixed regression model:

$$P_t = \beta\, Z_t + \theta_t + \varepsilon_t \qquad \text{(Eq. 3.2)}$$

where P_t denotes the concentration of BC, CO, NO, NO_2, NO_x, or O_3 measured during hour t and β is the coefficient of interest that describes the influence of factor Z_t on the hourly pollutant level. The factors assessed include time period of the day, temperature, wind speed, relative humidity, wind direction, weekend (Saturday and Sunday), and hourly traffic counts. The temporal factor was divided into four periods: morning rush hour (6 – 9am), mid-day (10am – 3pm), evening rush hour (4 – 8pm, used as reference group), late evening (9 – 24pm), and early morning (1 – 5am). The wind direction factor was divided into three directions: north (315 – 45 degrees), east (45 – 135 degrees, which leads to the monitoring sites being downwind of the highway), and south (135 – 225 degrees). θ_t represents time-specific random intercepts used to capture potential variations not explained by Z_t and ε_t represents residual random normal error. The regression relationship between pollutant concentrations and driving factors developed a simplified method compared to the use of chemical transport models or dispersion models. The multivariate regressions provide a direct relationship for health studies to better understand the driving factors for near-road exposures.

3.3. Results and Discussion

3.3.1 Observed near-road air pollutant concentrations

The NR DRIVE site and the NR GIT site measured CO, NO, NO_2, NOx, and BC concentration continuously from September 8, 2014 to January 5, 2015. The BC measured at the NR GIT site began sampling November 3, 2014. The NR DRIVE site also measured ozone (O_3), wind speed, and temperature. CO and NO_2 mean (standard deviation) concentrations at the NR DRIVE site were 425 ppb (210 ppb) and 29 ppb (15.5 ppb), respectively (Table 3.1). At the NR GIT site, average (standard deviation) concentrations measured for CO and NO_2 were 624 ppb (338 ppb) and 19.5 ppb (8.6 ppb), respectively (Table 3.1). In addition to the chemiluminescent method used to measure NO_2 at the NR DRIVE and NR GIT site, the NR DRIVE site measured NO_2 using the direct method with a mean (standard deviation) concentration of 22 ppb (11.7 ppb).

During the sampling period, CO and NO_2 hour maximums at both sites remained below the hourly national standards of 35 ppm for CO and 100 ppb for NO_2 (Figure 3.3) despite the prominent wind direction being from the east. The maximum hourly concentration at the NR DRIVE and NR GIT sites were 1860 ppb (CO) and 93.8 ppb (NO_2), and 2200 ppb (CO) and 51.6 ppb (NO_2), respectively (Table 3.1). Due to high concentrations that skewed the distribution causing a non-normal distribution, sites were compared using a Spearman's rank correlation. While the hourly time series show the two monitoring locations capturing the similar trends (Figure 3.3), the NR DRIVE site on average measured lower CO and NOx as well as higher NO_2 than the NR GIT location (Table 3.1). Temporal variability in CO and NO_2 hourly concentrations lead to a

Spearman's correlation of 0.18 (CO) and 0.72 (NO_2) (Figure 3.3). The average diurnal profiles also highlight the overall lower CO and NO_2 concentrations measured at the NR DRIVE site and the higher NO_2 concentrations measured at the NR GIT site (Figure 3.4).

Table 3.1 Hourly averages of NR DRIVE and NR GIT near-road continuous instrumentation. September 8, 2014 to January 5, 2015. BC: Black carbon, CO: Carbon monoxide, NO: Nitric oxide, NO2: Nitrogen oxide, NOx: Nitrogen oxides, O_3: Ozone, T: Temperature, N: Number of hours with observations, SD: Standard deviation, IQR: Inter Quartile Range, Min: Minimum observation, Max: Maximum observation

		BC (ug m^{-3})		CO (ppb)		NO (ppb)		NO_2 (ppb)		Direct NO_2 (ppb)	NOx (ppb)	
		DRIVE	GIT	DRIVE	GIT	DRIVE	GIT	DRIVE	GIT	DRIVE	DRIVE	GIT
Total 9/8-1/5	N	2282	1115	2178	2816	2666	2798	2666	2798	2878	2666	2798
	Mean	1.6	1.7	425	624	21	38	29	20	22	50	57
	SD	1.3	1.2	210	338	24	29	16	8.6	12	35	34
	IQR	0.7 - 2.2	0.86 - 2.2	278 - 515	400 - 800	5.6 - 29	17 - 51	17 - 38	13 - 25	13 - 29	25 - 67	32 - 74
	Min-Max	0.06 - 13	0.13 - 9.4	133 - 1860	0 - 2200	0 - 201	1.0 - 233	2.4 - 94	2.3 - 52	0 - 71	2.7 - 252	4.9 - 263
September 9/1-9/30	N	469	-	526	543	419	540	419	540	552	419	540
	Mean	1.9	-	414	689	18	34	32	19	23	50	52
	SD	1.2	-	157	264	15	20	13	6.4	8	26	24
	IQR	0.9 - 2.5	-	291 - 502	500 - 900	6.9 - 28	18 - 45	22 - 39	14 - 23	17 - 27	30 - 66	34 - 68
	Min-Max	0.2 - 8.8	-	136 - 1306	0 - 1500	0 - 95	1.3 - 118	4.4 - 82	4.8 - 42	7.0 - 55	3.7 - 174	11 - 139
October 10/1-10/31	N	529	-	672	713	663	720	663	720	743	663	720
	Mean	1.7	-	404	584	20	31	33	20	22	53	50
	SD	1.4	-	185	242	21	23	17	8.1	12	33	27
	IQR	0.7 - 2.3	-	275 - 498	400 - 700	5.9 - 26	14 - 42	19 - 44	13 - 25	17 - 30	27 - 71	30 - 66
	Min-Max	0.2 - 12	-	133 - 1304	0 - 1700	0.2 - 155	1.0 - 125	2.8 - 94	2.3 - 49	3.3 - 63	3.7 - 202	5.8 - 150
November 11/1-11/30	N	552	251	662	707	720	700	720	700	720	720	700
	Mean	1.4	1.4	430	665	20	38	29	21	22	49	59
	SD	1.3	1.1	227	356	25	33	17	11	15	39	40
	IQR	0.6 - 1.9	0.7 - 1.8	270 - 524	400 - 800	4.9 - 24	14 - 51	15 - 39	12 - 28	10 - 30	21 - 62	28 - 77
	Min-Max	0.2 - 13	0.2 - 5.8	138 - 1594	100 - 2100	0 - 180	1.0 - 186	4.2 - 82	2.8 - 52	0 - 70	4.0 - 225	4.9 - 213
December 12/1 - 12/31	N	612	744	318	733	744	722	744	722	743	744	722
	Mean	1.6	1.9	477	677	24	49	26	19	21	50	68
	SD	1.4	1.3	280	367	28	35	13	8	11	37	40
	IQR	0.7 - 2.0	1.0 - 2.4	285 - 593	500 - 900	5.5 - 34	23 - 65	16 - 34	12 - 24	13 - 27	24 - 67	39 - 89
	Min-Max	0.06 - 12	0.1 - 9.4	168 - 1860	0 - 2200	0.05 - 201	1 - 233	2.4 - 75	3.7 - 47	0.8 - 66	2.7 - 252	5.1 - 263

Table 3.1 (continued)

		Wind Speed (mph)	Temp (F)	Traffic Count
Total 9/8-1/5	N	2343	2343	1920
	Mean	2.4	56	12300
	SD	1.4	13	5780
	IQR	1.4 - 3.2	46 - 67	2180 - 19000
	Min-Max	0 - 7.0	23 - 86	1400 - 20700
September 9/1-9/30	N	226	226	288
	Mean	2.3	71	12600
	SD	1.0	6	5800
	IQR	1.6 - 3.0	67 - 74	7300 - 17300
	Min-Max	0 - 5.2	57 - 85	1560 - 20700
October 10/1-10/31	N	744	744	504
	Mean	2.0	66	12500
	SD	1.3	10	5800
	IQR	0.9 - 3.0	59 - 73	7500 - 17100
	Min-Max	0 - 6.0	40 - 86	1500 - 20200
November 11/1-11/30	N	676	676	456
	Mean	2.6	48	12100
	SD	1.6	11	5780
	IQR	1.4 - 3.6	41 - 57	6870 - 17080
	Min-Max	0 - 7.0	23 - 75	1400 - 20000
December 12/1 - 12/31	N	591	591	672
	Mean	2.5	49	12100
	SD	1.2	9	5780
	IQR	1.7 - 3.1	42 - 56	6680 - 17000
	Min-Max	0.04 - 6.9	30 - 72	1490 - 20600

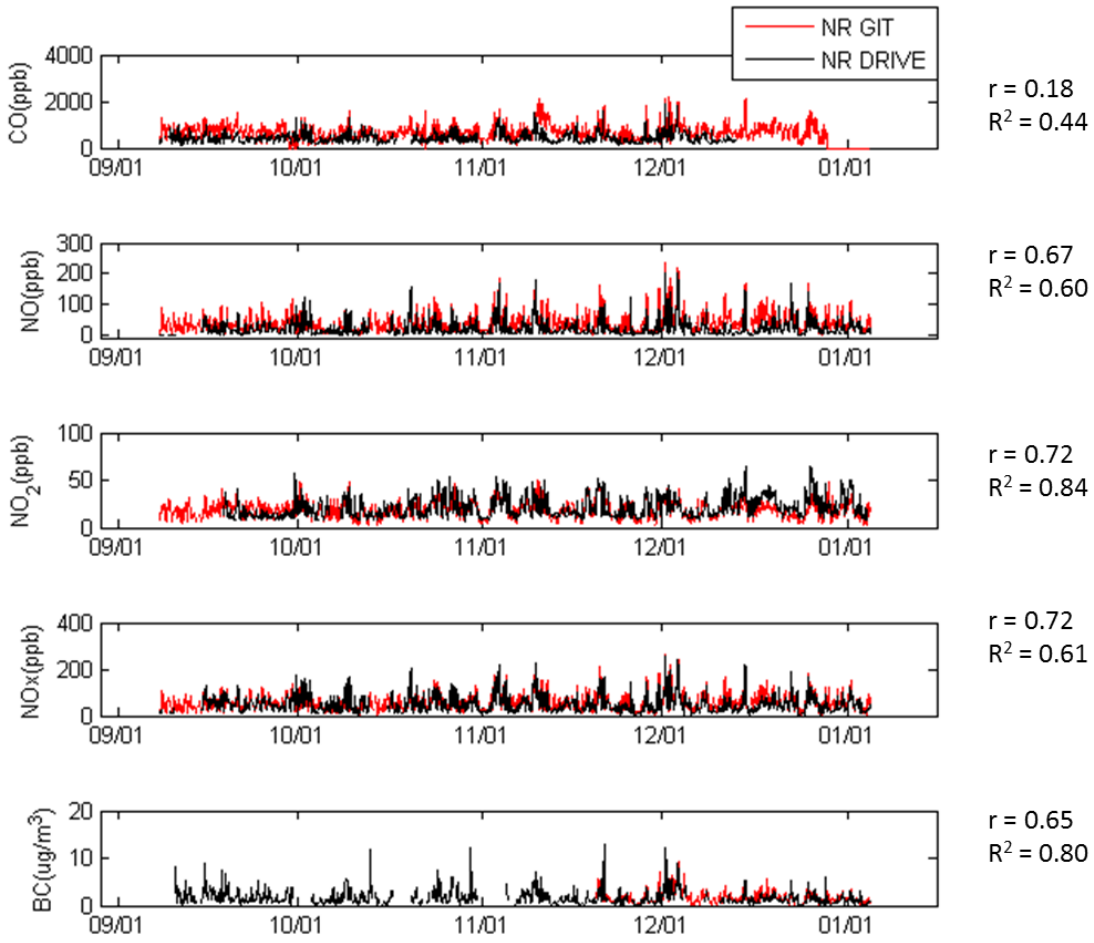

Figure 3.3 Time series of CO, NO, NO2, NOx, and BC at the NR DRIVE and the NR GIT site from September 8, 2014 to January 5, 2015

Less dispersion at the NR GIT site due to vegetation drove the bias between the NO and NO_2 concentrations at the NR DRIVE and NR GIT sites as well as different rates of mixing with ozone at the two sites. This is partly shown by the closer levels of NOx with the mean (standard deviation) concentration of 50 ppb (35 ppb) and 57 ppb (34 ppb) at the NR DRIVE and NR GIT sites, respectively. The NOx Spearman's correlation is 0.72 and consistent with the NO_2 correlation further suggestion a consistent bias drives the

differences in the NO_2 concentration (Figure 3.3). Both sites capture the morning and evening increase in TRAP concentrations in the same hour; however, one site consistently remains higher than the other throughout the average diurnal profile when comparing similar measurement methods for NO_2.

At the NR DRIVE and NR GIT sites, the mean (standard deviation) BC concentrations were 1.6 (1.3) ug m^{-3} and 1.7 (1.2) ug m^{-3}. While the NR GIT site only measured during November and December, similar monthly averaged concentrations were observed at both sites during these months. Diurnally, the BC concentration at both sites followed a similar trend with a morning peak from 9am to 11am and an afternoon minimum at 5pm (Figure 3.3). The BC diurnal trend mimicked the NO trend with an unbalanced bimodal distribution observing a maximum concentration in the morning 1.5 times greater than the evening peak. These pollutant distributions differed from the more balanced bimodal distributions of the CO and NO_2 diurnal profiles, which observed similar peak concentrations in the morning and evening.

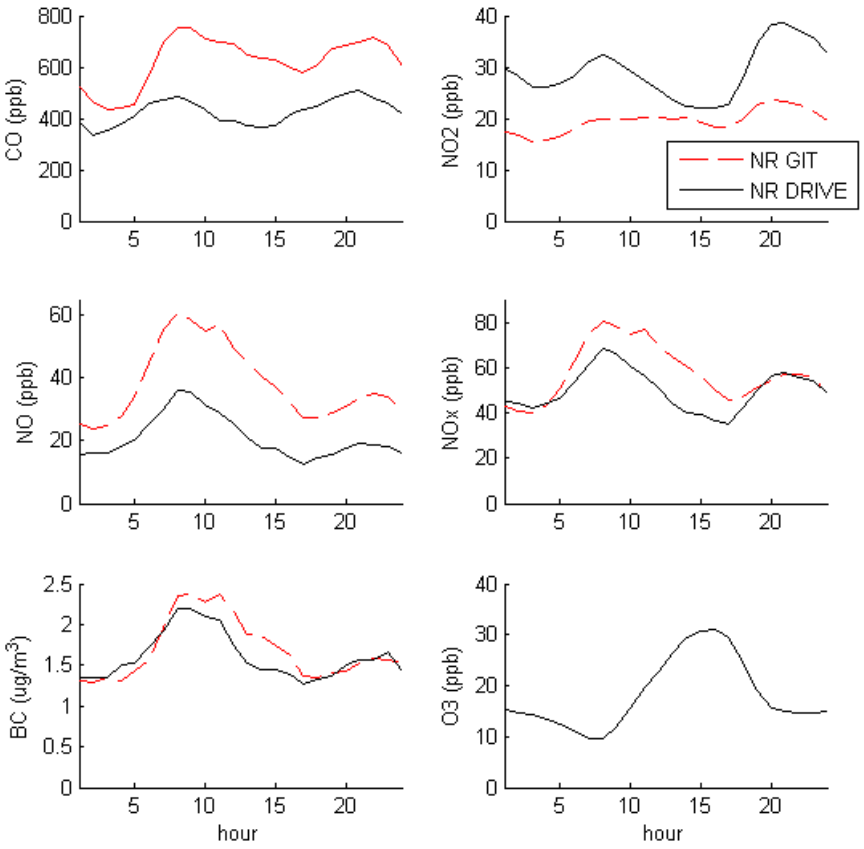

Figure 3.4 Diurnal profiles for CO, NO2, NO, NOx, BC, and O3 for the NR GIT and NR DRIVE sites from September 8, 2014 to January 5, 2015.

In order to better understand the impact vehicle emissions have on local concentrations, the near-road measurements were compared to the urban background and rural measurements around Atlanta, GA. Measurements for the urban background concentrations were collected at the highly instrumented, long term, Jefferson St. site, part of the Southeastern Aerosol Research and Characterization (SEARCH) network (Blanchard, Hidy, Tanenbaum, Edgerton, & Hartsell, 2013b; Edgerton et al., 2005, 2006; Hansen et al., 2006; Hansen et al., 2003; Liu et al., 2005; Solomon et al., 2003) located

2.3km west of the near-road sites. Average (maximum) hourly concentrations for urban background CO and NO_2 are 266 ppb (1732 ppb) and 12.6 ppb (94.4 ppb). Also part of the SEARCH network, the Yorkville site located about 40 miles northwest of Atlanta provides rural background pollutant concentrations. The average (maximum) hourly concentrations for rural CO and NO_2 are 175 ppb (524 ppb) and 2.2 ppb (26.9 ppb) respectively. Based on the difference in the means at the measurements sites from September 8, 2014 to January 5, 2015, the regional background contributes about 28% to the NR GIT site CO measurements, and the urban emissions contribute 15% to the NR GIT site CO measurements. The highway vehicle emissions are a significant source of the CO concentration measured in the near-road environment contributing the remaining 57% to the measured CO concentration at the NR GIT site. In contrast, the regional background is a smaller percentage of the NO_2 concentration measured in the near-road environment (11%) and the urban background contributed to about 53% of the NR GIT NO_2 measurements. These results suggest that localized regulations for CO would help reduce exposure to vehicle emissions since CO highway emissions are twice as high compared to the other regional and urban source. While CO levels are decreasing over the last decade, the biogenic component from oxidation of organic tree emissions is about 25% of the average contributing largely to the rural background. Alternatively, city scale regulations for NO_2 would help overall exposure since NO_2 highway emissions contribute only 35% to the near-road measurements.

Single pollutant concentrations measured in the near-road environment are commonly used as key indicators of the impact vehicle emissions have on local air quality within the near-road microenvironment. In order to assess the impacts from vehicle

emissions in the near-road environment, CO and NO_2 are commonly measured to represent primary vehicle emissions. The near-road measurements for both key TRAPs were elevated above the Atlanta urban and rural background concentrations suggesting traffic emissions contribute to elevated concentrations in the near-road environment. The NR DRIVE site concentrations for CO and NO_2 were 37% and 57% higher than the urban background concentrations. This is consistent with other EPA Near-road Monitoring Network sites in 2014, which reported mean NO_2 concentrations ranging from 9 to 24 ppb (DeWinter et al., 2018; EPA, 2016). While traffic emissions contribute to the elevated levels of primary pollutants in the near-road environment, other urban and region sources contribute a significant fraction affecting their use as tracers for vehicle emissions.

As part of the EPA Near-road Monitoring Network, there are two sites located in Atlanta. The NR GIT site in central Atlanta along highway I-75/I-85 with an AADT of 330,000 and the near-road South Dekalb (NR SDK) site located on the I-285 bypass around metro Atlanta with an AADT of about 140,000. The key pollutant measured at the NR GIT and NR SDK sites was NO_2 with mean (standard deviation) concentrations of 15.9 ppb (9.3 ppb) and 19.8 ppb (8.7 ppb), respectively. The correlation between the two sites was low ($r = 0.24$) and the diurnal profile shows the higher concentration at the NR GIT site occurring from early morning to evening (2am – 6pm) (Figure 3.6)

3.3.2 Assessment of traffic volume and meteorological factors impacting roadside concentrations

Traffic count and meteorological conditions are often key factors driving near-road TRAP concentrations. Average weekday traffic data during the sampling period on the

interstate demonstrates three peaks with the morning and evening rush hour events as well as a mid-day peak at about 2pm (Figure 3.5a). Instead of a common bimodal traffic count distribution (Richard Baldauf et al., 2012; Batterman, Cook, & Justin, 2015), this segment of highway has a consistently high traffic volume with vehicle counts rising quickly from 5am to 7am and more slowly until reaching a maximum at 3pm. Vehicle count slowly decreases from 3pm to 7pm and quickly drops reaching a minimum at 3am. The daily trend was consistent across all four months of the study with variability in the travel behavior based on weekday (Figure 3.5b) and weekend (Figure 3.5c). Traffic volume on the weekend displayed no peak in the morning or evening further highlighting the significance of both work commuting trips and the use of the highway for other daily trips.

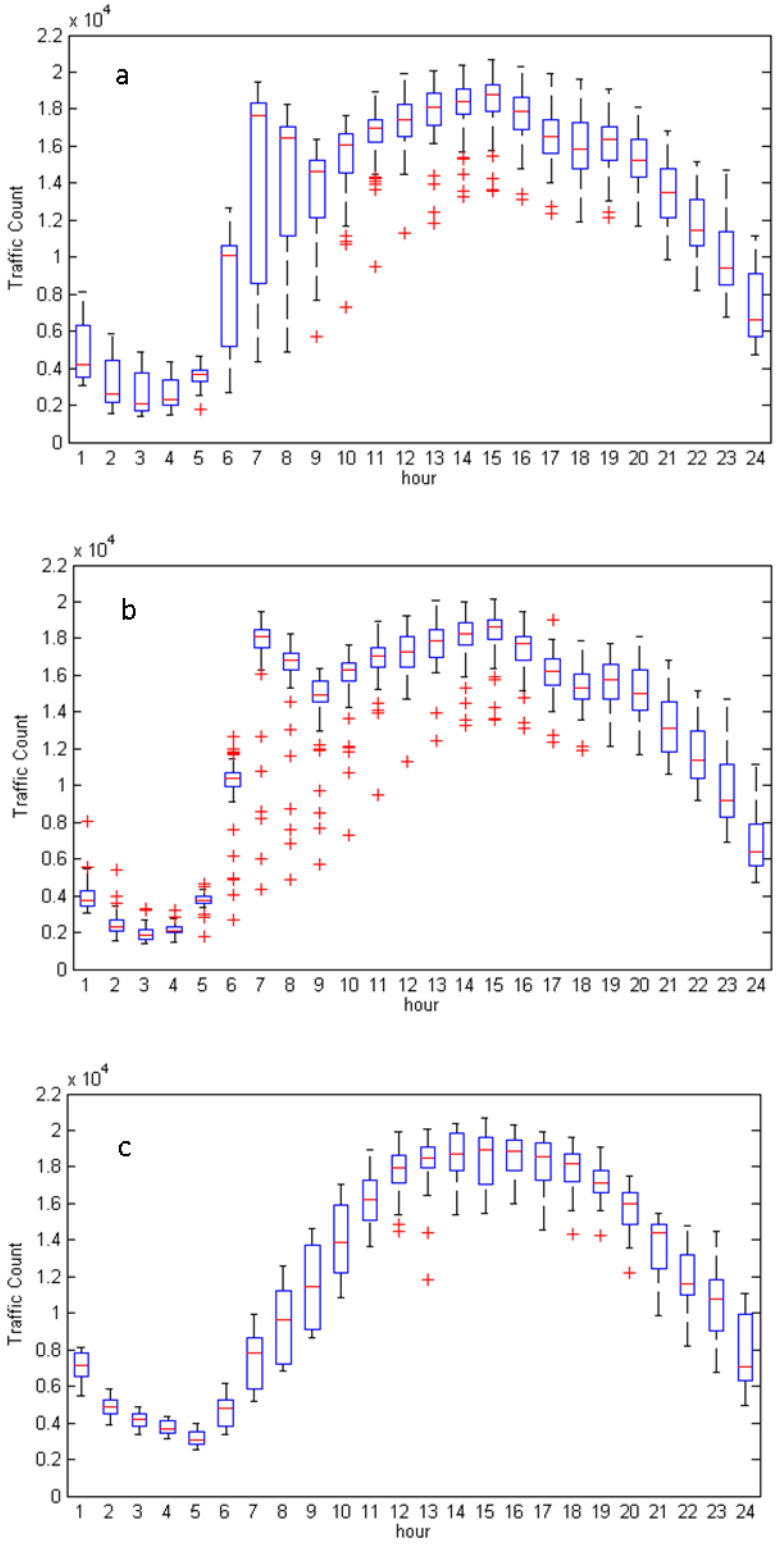

Figure 3.5 Hourly average traffic count of (a) total, (b) weekday, and (c) weekend variability from September 1, 2014 to December 31, 2014

The mean normalized diurnal profiles of the key TRAP species, vehicle counts, and meteorological conditions driving dispersion show the extent of daily variation for each pollutant and factor for the NR DRIVE site and the NR GIT site (Figure 3.6). The normalized CO, NO_2, and BC concentrations have a similar diurnal profile with a morning concentration peak at 10am, an evening peak at 10pm, and minimum concentrations observed at 3am and 4pm. The normalized hourly O_3 concentration measured at the NR DRIVE site had a maximum concentration at 6pm and a minimum at 10am. Since the minimum primary pollutant concentrations occur during the hours with maximum vehicle counts, this shows that highway traffic count alone is a poor indicator of diurnal pollutant levels.

The average vehicle speed and corresponding congestion patterns (Figure 3.6c) show highway traffic speeds remaining high throughout the night and reaching a minimum at 6pm during the evening rush hour period. As traffic counts rise at 6am, corresponding reductions in mean traffic speeds are observed from about 70mph to 40mph by 8am. While traffic counts remain high throughout the day, mean traffic speeds increase from 45mph at 10am before dropping to 15mph at 5pm. Since vehicle emissions rates remain fairly constant above 20mph (Barth, Scora, & Younglove) and traffic counts remain elevated throughout the day, diurnal emissions trends would suggest the highest vehicle emissions occur between 2pm and 8pm when measured species concentrations are lowest.

Mixing height data was generated by the WRF model for the 4km grid including the two central near-road locations, and varied diurnally. The mixing height remained low (about 260m) until about 7am, increased during the day until reaching a maximum height at about 3pm (typically about 1000m), and decreased until approximately 10pm. Mixing

height as well as ozone formation driven by photochemical activity resulted in a peak for both between 1pm and 6pm. As the mixing height increased in the morning, TRAP concentrations decrease reaching a minimum concentration at 3pm while the traffic count was reaching a maximum. By multiplying the concentration by the mixing height (Figure 3.6d and 3.6e), the increase mid-day when the mixing height is greatest shows the emissions increase with traffic count, but mixing height drives daily dynamics leading to minimum concentrations mid-day.

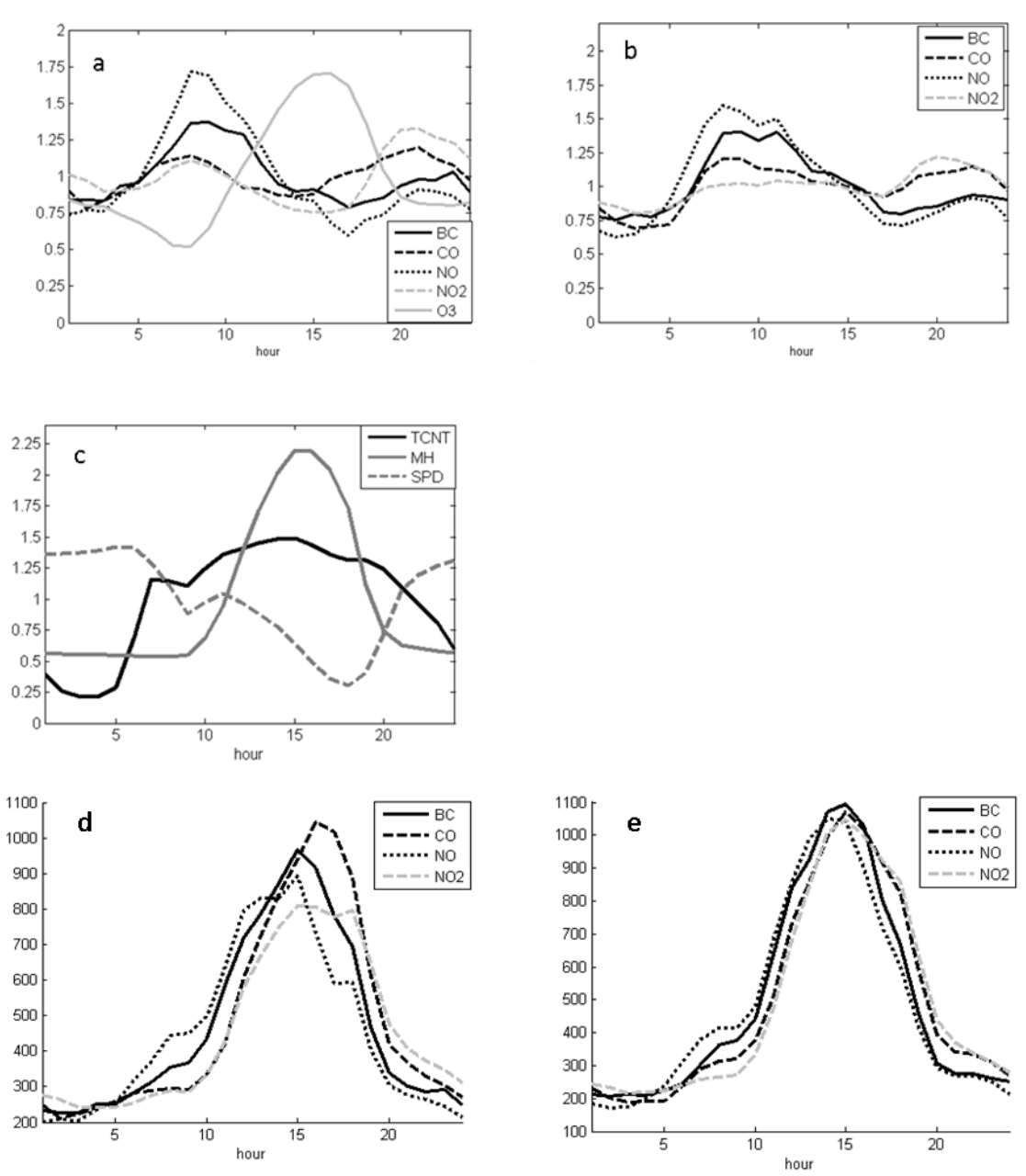

Figure 3.6 Hourly (a) NR DRIVE concentration data, (b) NR GIT concentration data, and (c) traffic parameter data normalized by mean. (d) NR DRIVE concentration normalized by mean and multiplied by mixing height. (e) NR GIT concentration normalized by mean and multiplied by mixing height. Data from September 8, 2014 to January 5, 2015. TCNT: Traffic count, MH: Mixing height, SPD: Traffic highway speed.

The two near-road sites along the same highway segment observed trends in primary TRAPs explained by rapid pollutant dispersion, particularly associated with the increase in convective mixing and increased wind speeds. Even at near-road sites, the diurnal convective mixing and wind speed has the dominant impact of TRAP concentrations (Figure 3.6). The high impact of meteorological factors compared to traffic count suggests a change in fate and transport properties affecting near-road concentration variability. This change impacts the applicability of traffic count as a mobile source tracers in quantifying exposure to traffic emissions. Further, an implication of the changing near-road environment is that future exposure studies aimed at characterizing health impacts of mobile emissions will need to consider different approaches for determining the mobile source contribution to ambient concentrations of single pollutants.

The pollutant diurnal patterns at the two near-road network sites were consistent with prior studies showing elevated concentrations of primary traffic pollutants occurring during morning rush hours when the atmospheric mixing is weak and emissions are high, then concentrations decreasing at the boundary layer increases. During the evening rush hours, the concentrations again increased and remain high throughout the night in spite of the greatly reduced emissions due to the low boundary layer. Both sites observed the typical bi-modal diurnal profile for the primary traffic-related air pollutants, but did not observe the same diurnal profile for vehicle count, which is often used as a proxy of exposure to traffic pollutants in health effect studies (Batterman, Cook, et al., 2015). Traffic counts on I-75/I-85 through metro Atlanta exhibited distinctive patterns of rising sharply in the early morning consistent with the beginning of the morning rush hour and reached a consistent peak vehicle count of approximately 20,000 vehicles per hour from 10am to 4pm. The

differences in the diurnal profile patterns between the vehicle counts and primary traffic pollutant concentrations highlights the predominate role meteorology and its influence on vertical dispersion have on the impact of traffic hotspots on adjacent areas.

The two EPA near-road monitors (NR GIT and NR SDK) were also compared to assess how exposure analyses may vary based on near-road monitoring locations in an urban area (Figure 3.7). While the time series for NO, NO_2, and NOx concentrations from January 1, 2015 to December 31, 2015 are within the same magnitude, the correlation of the two monitoring sites show a poor correlation ($R^2 = 0.4$) for the species. The comparison of the average diurnal profiles show different pollutant dynamics at the two sites. The NO and NO_2 concentrations are higher at the NR GIT site compared to the NR SDK site showing how total traffic count increases concentration in the near-road environment. While meteorology drove the diurnal profile observe at the NR GIT site, a traffic count double the observed AADT at the NR SDK site resulted in higher concentrations overall. At these sites, NO is likely more of an indicator of traffic tailpipe emissions with a concentration only higher during the hours with a higher traffic count. NO is directly emitted from tailpipes and NO_2 is a secondary pollutant that forms from NO emissions. NO_2 then remains high after sunset when photochemical reactions stop without sunlight. Monitoring site location characteristics, fleet composition, and traffic dynamics differed driving the diurnal profiles observed.

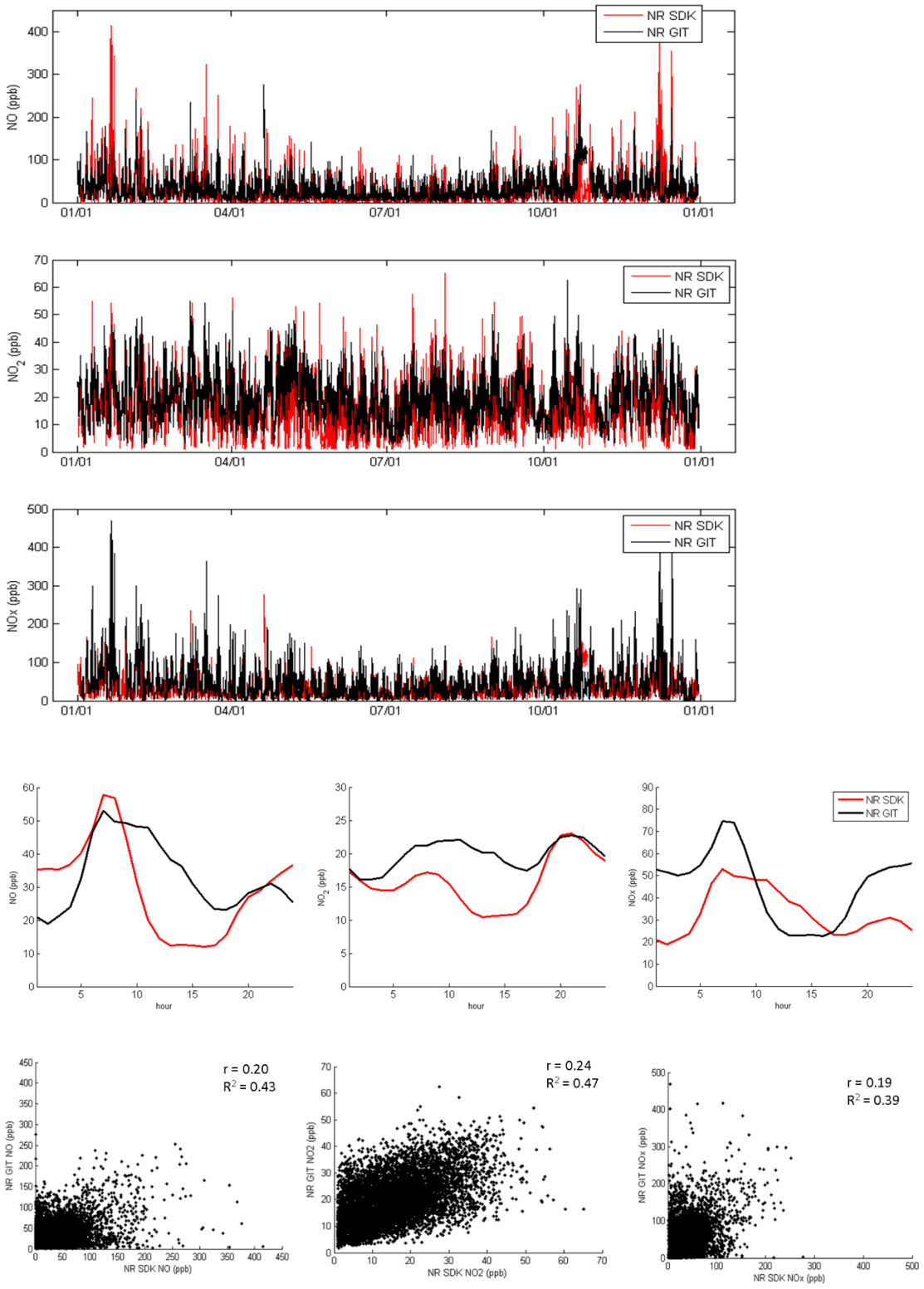

Figure 3.7 EPA Near-road Monitoring Network sites (NR GIT and NR SDK) in metro Atlanta for January 1, 2015 to December 31, 2015 for NO (ppb), NO2 (ppb), and NOx (ppb).

3.3.3 Assessment of combined traffic volume and meteorological factors leading to roadside concentrations

As part of the DRIVE study, exposures were modeled by integrating hourly concentrations. Further, the NAAQS for both NO_2 and CO have one-hour components. Thus, there is a need to link local and regional factors to hourly concentrations. This can be done using more complex dispersion modeling approaches or statistical methods. Statistical modeling can help identify if different factors influence the observations at the three near-road locations differently. Here, linear mixed modeling evaluated associations between pollutant concentrations and multiple possible contributing predictors to assess factors that drive the temporal variability observed at the different near-road sites. The regression coefficients for the models developed for the NR DRIVE site and the NR GIT site were compared to assess whether site differences along the same road segment can lead to significant differences in pollutant dynamics (Table 3.2). The regression coefficients for the two EPA near-road monitors (NR GIT and NR SDK) were also compared to assess how exposure analyses may vary based on near-road monitoring locations in an urban area (Table 3.3). Significance of a factor was determined by a p-value less than 0.05.

During the 2014 study period, the NR DRIVE site and the NR GIT site regression coefficients for BC and NO concentrations were positively associated with late evening to morning rush hour period (9pm to 9am) when the traffic count and mixing height were low and beginning to increase (Table 3.2). Also for both sites, wind direction and increasing wind speed were significantly associated with a decrease in all the primary pollutants (BC, CO, NO, NO_2, and NOx) indicative of dispersion away from the pollutant source.

Additionally, weekend days showed an association with a significant decreasing concentration for all pollutants (NO, NO_2, and BC) except CO. While many of the factors were significant for both sites, temperature was negatively significant for NO, NO2, and NOx only at the NR GIT site. Temperature was also significantly negative at the NR GIT site for BC and CO, while positively significant for the NR DRIVE site. The NR DRIVE site is located in an open parking lot indicative of increased photochemical reactions and pollutant dispersion compared to the NR GIT site, which was located within a tree barrier along the highway. The regression models also highlight the diminishing predictive power of traffic count on near-road pollutant levels. While the coefficient for traffic count is significant at both near-road sites and for all pollutants, the magnitude of the coefficient is low and therefore is not a key factor driving temporal variability of pollutant concentration.

For both EPA near-road monitoring network sites in 2015, increasing wind speed were significantly associated with a decrease in all the primary pollutants (BC, CO, NO, NO_2, and NOx) indicative of increased dispersion away from the highway sources (Table 3.3). Temperature was a significant factor to include in the model and was associated with a decrease in NO, NO_2, and NOx concentrations at the NR GIT, NR SDK, and SDK sites. While temperature was also associated with a decrease in BC and CO at the SDK and NR SDK sites respectively, temperature was associated with an increase in concentration at the NR GIT site. Weekend days were also associated with a significant decreasing concentration for NO, NO_2, and NOx at the NR GIT and NR SDK sites, however for BC, weekend days were a significant factor for increasing concentration at the NR GIT site and decreasing concentrations at the NR SDK site.

Table 3.2 Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations at the NR DRIVE and NR GIT sites from September 8, 2014 to January 5, 2015. *All covariates were included simultaneously in the model from each pollutant of interest. Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for BC: μg m^{-3}; Unit for CO, NO, NO_2, NOx, and O_3: ppb; *Significant with a P-value of 0.05

	NR DRIVE BC		NR GIT BC		NR DRIVE CO		NR GIT CO		NR DRIVE O_3	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-0am)	0.2*	(0.04, 0.42)	0.54*	(0.29, 0.79)	6.08	(-27.25, 39.41)	47.63*	(6.97, 88.29)	1.31*	(0.24, 2.38)
Early Morning (1-5am)	0.2*	(0.03, 0.44)	0.52*	(0.24, 0.79)	-8.52	(-44.92, 27.88)	-22.45	(-66.23, 21.34)	2.05*	(0.89, 3.20)
Morning Rush Hour (6-9am)	0.4*	(0.13, 0.64)	0.70*	(0.37, 1.03)	-11.98	(-55.88, 31.92)	-1.59	(-54.11, 50.94)	1.85*	(0.42, 3.27)
Mid Day (10am-3pm)	0.1	(-0.04, 0.32)	0.49*	(0.25, 0.72)	-27.31	(-58.23, 3.61)	25.87	(-11.83, 63.57)	1.54*	(0.56, 2.53)
Temperature	0.01*	(0.00, 0.02)	-0.02*	(-0.04, -0.00)	2.35*	(0.74, 3.97)	-4.00*	(-6.13, -1.86)	0.40*	(0.35, 0.45)
Relative Humidity	0.0	(0.00, 0.01)	0.00	(-0.01, 0.01)	-0.37	(-1.31, 0.58)	-0.33	(-1.59, 0.93)	-0.22*	(-0.26, -0.19)
Wind Speed	-0.2*	(-0.26, -0.14)	-0.19*	(-0.27, -0.12)	-40.75*	(-50.09, -31.40)	-45.35*	(-57.35, -33.34)	2.54*	(2.24, 2.84)
Northerly Wind	-0.3*	(-0.47, -0.08)	-0.74*	(-1.28, -0.20)	-36.78*	(-70.63, -2.92)	-90.25*	(-134.28, -46.23)	1.83*	(0.73, 2.93)
Easterly Wind	-0.7*	(-0.95, -0.51)	-1.25*	(-1.79, -0.70)	-59.43*	(-97.25, -21.61)	-191.16*	(-240.46, -141.87)	5.06*	(3.83, 6.29)
Southerly Wind	-0.3	(-0.76, 0.17)	-	-	-7.17	(-83.98, 69.64)	-222.118	(-324.75, -119.47)	3.44*	(0.83, 6.04)
Weekend	-0.6*	(-0.97, -0.26)	-0.81*	(-1.40, -0.22)	-11.84	(-74.45, 50.77)	-9.13	(-92.74, 74.49)	2.34*	(0.40, 4.28)
Traffic Count (per 1,000)	0.06*	(0.04, 0.07)	0.00*	(0.06, 0.09)	8.60*	(6.56, 10.64)	0.02*	(19.11, 24.09)	-0.31*	(-0.37, -0.24)
Mixing Heights (per 100 meters)	-0.03*	(-0.05, -0.01)	0.00	(-0.04, 0.02)	-3.41	(-7.12, 0.30)	-6.39*	(-11.14, -1.64)	0.47*	(0.06, 0.34)

	NR DRIVE NO		NR GIT NO		NR DRIVE NO_2		NR GIT NO_2		NR DIRVE NOx		NR GIT NOx	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-0am)	4.7*	(1.9, 8.7)	11.1*	(7.0, 15.3)	0.3	(-1.6, 2.2)	0.3	(-0.8, 1.4)	5.0*	(0.4, 9.7)	11.4*	(6.6, 16.2)
Early Morning (1-5am)	5.4*	(3.7, 12.5)	11.9*	(7.4, 16.5)	-2.2*	(-4.3, -0.2)	-1.0	(-2.2, 0.2)	3.2	(-1.8, 8.2)	10.9*	(5.6, 16.1)
Morning Rush Hour (6-9am)	7.5*	(-0.3, 6.0)	14.6*	(9.2, 20.0)	-1.9	(-4.3, 0.6)	-0.4	(-1.8, 1.1)	5.7	(-0.3, 11.6)	14.2*	(7.9, 20.4)
Mid Day (10am-3pm)	3.3*	(2.1, 9.5)	7.8*	(4.0, 11.7)	-2.0*	(-3.7, -0.2)	0.2	(-0.8, 1.2)	1.3	(-3.0, 5.6)	7.9*	(3.5, 12.4)
Temperature	-0.1	(-0.3, 0.04)	-0.8*	(-1.0, -0.6)	0.04	(-0.05, 0.1)	-0.05*	(-0.1, -0.0)	-0.06	(-0.3, 0.2)	-0.9*	(-1.1, -0.7)
Relative Humidity	0.2*	(0.1, 0.3)	0.1*	(0.01, 0.2)	-0.2*	(-0.2, -0.1)	-0.1*	(-0.2, -0.2)	0.06	(-0.07, 0.2)	0.0	(-0.1, 0.1)
Wind Speed	-1.8*	(-2.9, -1.0)	-3.0*	(-4.2, -1.8)	-5.1*	(-5.6, -4.5)	-2.4*	(-2.7, -2.1)	-6.9*	(-8.2, -5.6)	-5.3*	(-6.7, -4.0)
Northerly Wind	-5.6*	(-29.4, -12.7)	-9.2*	(-13.6, -4.8)	-4.4*	(-6.4, -2.4)	-3.3*	(-4.5, -2.2)	-10.0*	(-14.9, -5.2)	-12.5*	(-17.5, -7.4)
Easterly Wind	-19.0*	(-23.2, -15.2)	-19.7*	(-24.6, -14.8)	-10.2*	(-12.4, -8.0)	-5.9*	(-7.2, -4.6)	-29.3*	(-34.7, -23.9)	-25.5*	(-31.2, -19.9)
Southerly Wind	-20.7*	(-9.3, -2.1)	-26.6*	(-36.8, -16.4)	-3.2*	(-7.8, 1.4)	-3.0*	(-5.7, -0.3)	-24.0*	(-35.2, -12.7)	-29.5*	(-41.4, -17.7)
Weekend	-9.1*	(-14.8, -3.2)	-13.1*	(-20.3, -5.9)	-5.0*	(-8.1, -1.8)	-2.4*	(-4.4, -0.5)	-14.0*	(-22.0, -6.1)	-15.4*	(-24.0, -6.8)
Traffic Count (per 1,000)	1.1*	(0.9, 1.3)	0.0*	(2.1, 2.6)	0.5*	(0.4, 0.6)	0.0*	(0.4, 0.5)	1.6*	(1.3, 1.9)	2.8*	(2.5, 3.1)
Mixing Heights (per 100 meters)	-0.4	(-0.8, 0.05)	0.0	(-0.8, 0.2)	-0.6*	(-0.9, -0.4)	0.0*	(-0.5, -0.2)	-1.0*	(-1.5, -0.5)	-0.6*	(-1.2, -0.08)

Table 3.3 Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations at the NR GIT, SKD, and NR SDK sites from January 1, 2015 to December 31, 2015. All covariates were included simultaneously in the model from each pollutant of interest. Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for BC: μg m^{-3}; Unit for CO, NO, NO_2, NOx, and O_3: ppb; *Significant with a P-value of 0.05

	NR GIT NO		SDK NO		NR SDK NO		NR GIT NO_2		SDK NO2		NR SDK NO2	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-0am)	8.4*	(6.4, 10.3)	5.8*	(4.1, 7.4)	2.9*	(0.8, 5.0)	2.4*	(1.68, 3.1)	1.7*	(1.2, 2.2)	-1.5*	(-2.1, -1.0)
Early Morning (1-5am)	12.8*	(10.7, 15.0)	7.4*	(5.9, 9.0)	2.2*	(0.2, 4.2)	0.9*	(0.15, 1.7)	-1.8*	(-2.2, -1.3)	-5.6*	(-6.2, -5.1)
Morning Rush Hour (6-9am)	17.6*	(15.0, 20.1)	5.9*	(3.7, 8.0)	3.7*	(1.0, 6.4)	2.7*	(1.75, 3.6)	-0.2	(-0.8, 0.4)	-3.0*	(-3.8, -2.3)
Mid Day (10am-3pm)	3.5	(1.7, 5.3)	6.2*	(4.7, 7.8)	2.9*	(0.9, 4.8)	-0.3	(-0.98, 0.3)	-0.6*	(-1.1, -0.1)	-3.4*	(-4.0, -2.9)
Temperature	-0.5*	(-0.6, -0.4)	-1.1*	(-1.2, -1.1)	-1.3*	(-1.4, -1.2)	0.04	(-0.0, 0.1)	-0.2*	(-0.2, -0.2)	-0.2*	(-0.2, -0.1)
Relative Humidity	0.1*	(0.1, 0.2)	0.02*	(0.0, 0.04)	0.01	(-0.02, 0.03)	0.04*	(0.02, 0.05)	0.02*	(0.01, 0.02)	0.0	(0.0, 0.01)
Wind Speed	2.3*	(1.5, 3.0)	-4.3*	(-5.0, -3.6)	-8.7*	(-9.5, -7.8)	-0.8*	(-1.1, -0.5)	-2.5*	(-2.7, -2.3)	-3.3*	(-3.6, -3.1)
Northerly Wind	-3.5*	(-5.8, -1.3)	0.4	(-1.0, 1.7)	0.2	(-1.5, 1.9)	-0.3	(-1.1, 0.6)	-1.05*	(-1.5, -0.6)	-2.0*	(-2.5, -1.5)
Easterly Wind	8.0*	(5.9, 10.2)	-4.6*	(-5.8, -3.4)	0.8	(-0.7, 2.4)	3.5*	(2.7, 4.3)	-1.4*	(-1.8, -1.1)	1.0*	(0.5, 1.4)
Southerly Wind	8.7*	(7.3, 10.1)	-3.02*	(-4.0, -2.0)	2.2*	(0.9, 3.5)	3.8*	(3.3, 4.3)	0.2	(-0.2, 0.5)	1.7*	(1.3, 2.1)
Weekend	-11.4*	(-14.9, -7.9)	-6.3*	(-11.2, -1.4)	-14.8*	(-19.4, -10.3)	-3.0*	(-4.8, -1.1)	-1.3	(-2.2, -0.3)	-3.3*	(-4.4, -2.3)
Traffic Count (per 1,000)	1.7*	(1.6, 1.8)	-	-	-	-	0.4*	(0.4, 0.5)	-	-	-	-
Mixing Heights (per 100 meters)	-	-	-	-	-	-	-	-	-	-	-	-

	NR GIT NOx		SDK NOx		NR SDK NOx	
	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-0am)	10.83*	(8.52 , 13.15)	7.13*	(5.35 , 8.91)	1.11	(-1.21 , 3.42)
Early Morning (1-5am)	13.99*	(11.43 , 16.55)	5.39*	(3.69 , 7.10)	-3.50*	(-5.70 , -1.29)
Morning Rush Hour (6-9am)	20.30*	(17.31 , 23.28)	5.24*	(2.87 , 7.60)	0.71	(-2.29 , 3.71)
Mid Day (10am-3pm)	2.94*	(0.78 , 5.09)	5.76*	(4.08 , 7.45)	-0.28	(-2.48 , 1.92)
Temperature	-0.48*	(-0.61 , -0.35)	-1.33*	(-1.42 , -1.25)	-1.41*	(-1.50 , -1.31)
Relative Humidity	0.15*	(0.10 , 0.19)	0.04*	(0.02 , 0.06)	0.01	(-0.01 , 0.04)
Wind Speed	1.39*	(0.48 , 2.30)	-6.72*	(-7.46 , -5.99)	-12.06*	(-13.00 , -11.11)
Northerly Wind	-4.41*	(-7.12 , -1.71)	-0.58	(-2.07 , 0.91)	-1.73	(-3.65 , 0.20)
Easterly Wind	11.51*	(8.92 , 14.10)	-5.76*	(-7.06 , -4.45)	1.81*	(0.11 , 3.52)
Southerly Wind	12.61*	(10.96 , 14.26)	-2.85*	(-3.96 , -1.74)	3.83*	(2.36 , 5.31)
Weekend	-13.91*	(-18.45 , -9.37)	-7.46*	(-12.83 , -2.09)	-18.15*	(-23.08 , -13.23)
Traffic Count (per 1,000)	2.13*	(1.97, 2.29)	-	-	-	-
Mixing Heights (per 100 meters)	-	-	-	-	-	-

Table 3.3 (continued)

	NR GIT BC		NR SDK BC		NR GIT CO		SDK CO	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-0am)	-1.11*	(-2.22 , 0.00)	0.08	(-0.04 , 0.21)	83.64*	(62.03 , 105.25)	41.99*	(31.83 , 52.14)
Early Morning (1-5am)	0.96*	(0.77 , 1.14)	-0.01	(-0.12 , 0.11)	55.35*	(31.65 , 79.05)	16.82*	(6.83 , 26.80)
Morning Rush Hour (6-9am)	1.50*	(1.30 , 1.70)	0.38*	(0.22 , 0.53)	81.07*	(53.44 , 108.69)	26.41*	(12.79 , 40.03)
Mid Day (10am-3pm)	2.03*	(1.79 , 2.27)	0.07	(-0.04 , 0.19)	-29.89*	(-49.95 , -9.82)	21.89*	(12.18 , 31.61)
Temperature	0.57*	(0.40 , 0.74)	-0.04*	(-0.05 , -0.04)	2.30*	(0.79 , 3.82)	-7.95*	(-8.45 , -7.46)
Relative Humidity	0.01	(0.00 , 0.02)	0.00	(0.00 , 0.00)	3.20*	(2.74 , 3.66)	0.26*	(0.13 , 0.39)
Wind Speed	0.00*	(0.00 , 0.01)	-0.38*	(-0.43 , -0.32)	11.07*	(2.39 , 19.76)	-48.15*	(-52.53 , -43.78)
Northerly Wind	0.27	(0.19 , 0.35)	-0.08	(-0.19 , 0.02)	-41.72*	(-67.80 , -15.64)	-5.39	(-13.97 , 3.18)
Easterly Wind	-0.16*	(-0.42 , 0.09)	0.06	(-0.03 , 0.15)	119.79*	(94.62 , 144.96)	-29.55*	(-37.22 , -21.87)
Southerly Wind	0.50*	(0.30 , 0.70)	0.12*	(0.04 , 0.20)	122.78*	(106.84 , 138.72)	-12.56*	(-19.07 , -6.05)
Weekend	0.43*	(0.31 , 0.56)	-1.04*	(-1.36 , -0.71)	-16.08	(-75.81 , 43.66)	8.90	(-24.31 , 42.11)
Traffic Count (per 1,000)	-1.61*	(-1.88 , -1.35)	-	-	0.02*	(0.02 , 0.03)	-	-
Mixing Heights (per 100 meters)	-	-	-	-	-	-	-	-

3.3.4 Changing Near-road Environment

The two near-road monitoring locations in Atlanta as part of the EPA Near-road Monitoring Network do not provide long-term measurements to understand how the near-road environment has changed over the last decade. A state operated, routine monitoring site (SDK), while not associated with the EPA Near-Road Monitoring Network, provides a long-term concentration data set located within 650m of the I-285 bypass around metro Atlanta with an AADT of about 140,000. The hourly average (maximum) concentrations from September 8, 2014 to January 5, 2015 for CO and NO_2 were 323 ppb (1259 ppb) and 9.7 ppb (57.5 ppb) respectively. From 2000 to 2010, the observed CO and NO_2 concentration at the SDK site decreased 33% (496 to 345 ppb) and 42% (18.1 to 13.7 ppb), respectively. For the same period from 2000 to 2010, the CO concentration decrease was 50% (561 to 282 ppb) and 9% (180 to 163 ppb) at the urban (JST) and rural (YRK) background sites. Similarly, the NO_2 concentration decrease was 30% (21.9 to 15.3 ppb) and 60% (5.4 to 2.2 ppb), respectively. Decreased mobile emissions have contributed to a decrease in near-road concentrations for primary traffic-related air pollutants; however, a decrease in overall concentrations locally and regionally have also contributed to the decrease in the near-road environment. Further, the difference in the rate of decrease from sources contributes to the changing near-road environment and how to characterize near-road exposure.

As observed here and elsewhere, near-road TRAP concentrations are less elevated above background levels than prior near-road studies around the country (Beckerman et al., 2008; S. E. Sarnat et al., 2008) suggesting that future studies will need to consider different approaches for characterizing the spatial gradients and exposures to mobile

sources. Initial results from the EPA Near-road Monitoring Network support these results. Of the 61 near-road sites active in 2015, only five hourly concentrations exceeded the hourly NO_2 standard of 100 ppb and at all the sites the 98th percentile of the daily 1-hour maximum was below the standard (DeWinter et al., 2018). The NO_2 concentration average across the country ranged from about 9 ppb to 30 ppb.

Nationally, estimates of on-road mobile source emissions of NOx have decreased about 50% since 2004 and emissions of CO show that 2014 levels are about 49% of those in 2004 and 25% of those in 1994 (Dallmann & Harley, 2010). In the metro Atlanta area, long term analysis data from the urban background site part of the SEARCH network shows that CO, NOx, and BC levels have decreased by 350 ppb (7.2% per year), 35 ppb (7.3% per year), and 1.25 μg m^{-3}(6.8% per year), respectively, from 1999 to 2011. Source apportionment analysis at this site indicated mobile source-related PM2.5 decreased by about half during the same period (Blanchard, Tanenbaum, et al., 2013). These declining trends are expected to continue in the future as fleet turnover to newer vehicles continues, new mobile source emissions controls are introduced and additional policy interventions are implemented.

3.4 Conclusion

Elevated traffic-related air pollutants are linked to adverse health effects and often health analysis rely on only a few monitoring locations to quantify exposure of individuals in the near-road environment. Comparison of two near-road monitors along the same highway segment showed site-to-site differences influencing pollutant concentrations. A single near-road monitor can characterize the pollutant temporal variability and assess the

concentration driving factors for analyzing exposure. Site characteristics can contribute to localized concentrations and this affect was observed when comparing the two near-road sites in this study. The two sites were 300m apart and both within a meter of a heavily trafficked highway. While the two sites observed similar diurnal and temporal variability, site differences including increased vegetation around one location reduced dispersion in comparison to the location by an open, asphalt parking lot. Reduced dispersion lead to higher CO and NO concentrations and lower NO_2 concentrations over the study period. These site differences contributed to different pollutant dynamics along the same highway segment highlighting the important of site location. The near-road pollutant concentrations measured show a reduced impact from the highway sources with levels less elevated above background concentrations than in prior studies, consistent with other studies throughout North America. These decreased concentrations indicate the effectiveness of mobile source emission controls leading to a decreased relative contribution from vehicles to urban air pollution. This finding indicates a changing near-road environment that will affect future approaches to characterizing vehicle emission hotspots and their impacts on exposures.

CHAPTER 4

EVALUATING MULTIPOLLUTANT AIR QUALITY METRICS FOR USE IN CHARACTERIZING HEALTH-RELATED EXPOSURES IN THE NEAR-ROAD ENVIRONMENT

Vehicular emissions are linked to several adverse health effects; however, quantifying exposure to the single source is increasingly difficult as vehicle emissions continue to decrease. As part of a larger study aimed to understand exposure of individuals in the near-road environment, an intensive measurement campaign collected traffic-related air pollutants (TRAPs) and related data (e.g., meteorology, traffic, regional air pollutant levels) in Atlanta along one of the busiest highway corridors in the US. Given the complexity of the near-road environment, multipollutant exposure metrics based on statistical emissions-based ratios or a metric of particulate matter toxicity tracers were characterized and compared to traditional single species methods for assessing exposure to mobile source emissions. The statistical emissions-based ratio metric provided a more spatially stable metrics for exposure, though varying based on location, the metric had inconsistent results among the six sites within a kilometer of the highway. The metric of particle toxicity based on oxidative potential was not a good indicator of primary vehicle emissions as it was more related to secondary pollutants from atmospheric processing.

4.1. Introduction

On-road vehicles lead to elevated concentrations of traffic-related air pollutants (TRAPs) including nitrogen oxides (NOx), volatile organic compounds (VOCs), carbon monoxide (CO), and primary fine particulate matter ($PM_{2.5}$) within the near-road microenvironment. As vehicle emissions continue to decrease with improved engine technologies and associated emissions control systems, and fuel regulations, the near-road concentration of TRAPs have also decreased nationally, though vehicle emissions remain a significant source for TRAPs in urban areas. Over the last two decades, the per-vehicle emissions reductions out-pace increases in vehicle miles travelled (VMT) and measured near-road concentrations have decreased in the last couple decades (Blanchard, Tanenbaum, & Hidy, 2012). This trend is expected to continue, though flatten, in the future.

In many studies, single species TRAP have been linked to a range of acute and chronic adverse health effects (see (HEI, 2009c) and references there in). Epidemiologic studies commonly utilize single-species tracers or proxies for understanding the health effects of traffic-related emissions (Brook et al., 2007; Hoek, Brunekreef, Goldbohm, Fischer, & van den Brandt, 2002; Levy, Mihele, Lu, Narayan, & Brook, 2014). TRAP concentrations, however, can vary dramatically due to the large spatial gradients that exist in the near-road environment (R. Baldauf et al., 2009). Gradients vary by pollutant as well as temporally throughout the day (D. H. Liang et al., 2018). Synergistic effects among air pollutants cause measureable health effects (Mauderly & Samet, 2009). As vehicle emissions decrease, the elevated concentration at the roadway decreases making it more difficult to distinguish the local emissions source from the background concentration (Ayala et al., 2012). Multipollutant approaches, including the use of source apportioned

measures of primary traffic pollution emissions, have been used to consider health risks from combined exposures to traffic mixtures (N. A. Janssen et al., 2011; M. M. Oakes et al., 2014; Ostro et al., 2007; Pachon et al., 2012).

4.1.1 Multipollutant perspective

Recent regulatory intervention has shifted towards adopting a multipollutant perspective for sources with many emitted pollutants to maximize the benefits of control expenditures and minimize population and ecosystem exposure (Council, 2004; Dominici, Peng, Barr, & Bell, 2010; Vedal, 2011). However, in order to transition to more multipollutant air quality regulations, improvements are needed to understand how well multipollutant regulations reduce exposure (Hidy & Pennell, 2010). In addition to setting multipollutant regulations, further development and assessment of statistical methods are needed for understanding the benefits of using multipollutant exposure metrics (HEI, 2009b) and assessing health impacts of mixtures. While populations are exposed to multiple pollutants simultaneously, which encourages shifting to a multipollutant approach, several different aspects need to be considered in the assessment of multipollutant metrics (Greenbaum & Shaikh, 2010; Mauderly et al., 2010).

For highly heterogeneous sources, a multipollutant framework provides new opportunities to characterize source emissions or biologically-relevant exposures. While a wide range of multipollutant metrics has been developed, each has benefits and limitations to assessing source impacts and the related health outcomes (Oakes, Baxter, & Long, 2014). Single tracer pollutants are easy to use and measure, but are emitted by several sources and may not be specific to a single source. Emissions-based indicators are simple

to calculate using weighted average concentrations based on source contribution, but are limited based on the accuracy of emissions inventories. The Integrated Mobile Source Indicator (IMSI) is an emissions-based, multipollutant metric derived from elemental carbon, carbon monoxide, and nitrogen oxide concentrations, along with the fraction of these species emitted by vehicles, to develop integrated estimates of vehicles impacts (Pachon et al., 2012). Developed using ambient concentrations in Atlanta, the IMSI has also been applied to two other cities with different emissions profiles to show applicability in a range of city types (M. M. Oakes et al., 2014).

Biologically-relevant metrics include a range of cellular and acellular assays used to quantify oxidative potential of $PM_{2.5}$ (Ayres et al., 2008; Charrier et al., 2015; Fang et al., 2016; N. A. H. Janssen et al., 2015). The acellular, dithiothreitol (DTT) assay is a cumulative method used to characterize $PM_{2.5}$ exposure through its ability to elicit reactive oxygen species (ROS) (Ghio et al., 2012; N. Li et al., 2003; Squadrito et al., 2001; Tao et al., 2003). Several studies have found an association between measures of oxidative potential and various health outcomes and some studies have found a stronger association of oxidative potential with health effects than $PM_{2.5}$ concentration (Bates et al., 2015; Bilenko et al., 2015; Dadvand et al., 2014; Steenhof et al., 2011; Steenhof et al., 2013; Yang et al., 2016). Not only has the DTT assay shown an association with respiratory and cardiovascular health outcomes, the assay is more sensitive to organic compounds compared to other acellular assays (Fang et al., 2016). Laboratory studies have found mobile source emissions are DTT-active (Q. F. Li, Wyatt, & Kamens, 2009; McWhinney et al., 2013; McWhinney et al., 2011). Previous studies have also quantified oxidative potential of vehicle emissions directly considering exhaust emissions and tire wear (Biswas

et al., 2009; Boogaard et al., 2012; Cheung et al., 2010; Gao, Fang, Verma, Zeng, & Weber, 2017; N. A. H. Janssen et al., 2014; Kelly, 2011; Q. F. Li et al., 2009; McWhinney et al., 2013; Yang et al., 2015). While primary vehicle emissions show consistent contribution throughout the year, secondary oxidation processes drive ROS potential in the summer and biomass burning drives the winter potential in the southeastern United States (Verma et al., 2014). While vehicle tailpipe emissions have decreased in the last decade, non-tailpipe emissions are a greater contribution to the oxidative potential of mobile emissions (Shirmohammadi et al., 2016; Shirmohammadi et al., 2017). Previous studies therefore suggest that the DTT assay may constitute an alternative indicator for characterizing exposure to mobile source emissions in the near-road environment.

4.1.2 Changing Near-road Environment

There is growing evidence that the near-road environment is changing rapidly and that traditional source contributions, fate and transport properties, and exposure factors already differ from those reported previously in existing literature (Blanchard, Hidy, Tanenbaum, Edgerton, & Hartsell, 2013a; Blanchard, Hidy, et al., 2013b; Blanchard, Tanenbaum, et al., 2013; Henneman et al., 2015; Vijayaraghavan et al., 2014). Substantial gaps exist in our understanding of how both traditional and non-traditional metrics vary spatially and temporally in the near-road environment, how they compare with each other, and whether they offer accurate means of assigning exposures to primary traffic emissions.

The EPA Near-road Monitoring Network was established in 2014 to improve population exposure assessments for individuals in the near-road environment. Chosen based on population and annual average daily traffic (AADT) count, Atlanta, GA is one of

the 54 cities across the country participating in the network system (DeWinter et al., 2018). To assess factors affecting exposure in the near-road environment, the Dorm Room Inhalation to Vehicle Exhaust (DRIVE) study was conducted in Atlanta, GA to characterize the spatial and temporal distribution of TRAPs along one of the busiest highway corridors in the US. A focus of the study was to characterize factors driving the pollutant concentration variability and to assess integrated traffic exposure metrics for applications in epidemiological studies (D. H. Liang et al., 2018).

This study aims to characterize the variability in a multipollutant exposure indicator that utilized the measurements collected during the DRIVE study. The emissions-based metric utilizes the national emissions inventory source ratios for primary, easily-measured mobile emissions to provide accurate estimates of short-term changes in exposures to traffic-related air pollution for use in acute health impact studies without the need for source apportionment (M. Oakes et al., 2014; Pachon et al., 2012). The biologically-based, acellular assay uses filter-based PM2.5 measurements to provide estimates of oxidative potential at multiple sites within a kilometer of a highly trafficked highway. The metrics were spatially and temporally compared to individual TRAPs measured at sites with varying distances from a major highway. The dynamics of the metrics were also assessed in relationship to local meteorological and traffic conditions. Understanding how well a multipollutant metric represents exposure to vehicle emissions based on the location of TRAP measurements has important implications for future regulatory and health assessment frameworks and the use of multipollutant indicators.

4.2. Methods

The DRIVE study focuses on pollutant dynamics and exposures in the near-road environment specifically considering the area around a section of heavily trafficked interstate in urban Atlanta. The study included three monitoring locations on the Georgia Institute of Technology (GIT) campus in Atlanta, GA that monitor gaseous and aerosol traffic-related air pollutant concentrations at a distance of 3m, 60m, and 1.4km from the highway. The latter two sites included indoor monitoring to assess infiltration rates and include indoor exposures. Measurements of continuous TRAPs included carbon monoxide (CO), nitrogen oxides (NOx), and particulate black carbon (BC) and integrated daily $PM_{2.5}$ were collected from September 8, 2014 to January 5, 2015. The overall objective of the DRIVE study was to capture exposures to traffic emission on the campus as students were being monitored for personal exposures and biomarker impacts. A more detailed analysis focusing on near-road monitoring and spatial gradients observed in this near-road environmental are discussed elsewhere (D. Liang et al., 2018; D. H. Liang et al., 2018). Additional measurements from an urban background site and a site part of the EPA Near-road Monitoring Network were included in this multipollutant analysis.

4.2.1 Site description and instrumentation

The DRIVE study focuses on emissions and related exposures from a segment of arterial interstate where Interstate 75 and Interstate 85 (I-75/I-85) have merged in the center of Atlanta, Georgia. During the study period, this highway segment supported an annual average daily traffic (AADT) count of about 330,000 vehicles of primarily light-duty

gasoline passenger cars and trucks. Heavy-duty diesel trucks made up approximately four percent of the total daily vehicles on this portion of the highway.

The DRIVE study sampling locations included a near-road site and two sites located in the main dormitory clusters on the GIT campus (Figure 4.1). The campus occupies the land up to 1.5km west of the highway and contains limited vehicle access roads. The near-road sampling site (NR DRIVE) was located in a parking lot with less than 85 passenger vehicle spots located about 5m from the west side of the fifteen-lane highway (eight southbound and seven northbound) to the south of 10th Street and to the north of North Avenue. The vertical height from the road to the surrounding land was 0.5 meters on the west side of the highway. Surface streets on the east side of the highway follow a gridded pattern with an average block length of 140m and an AADT 15 times less than that of the highway. The CO (Thermo Model 48i), NOx (Teledyne API 200A), and BC (Magee Aethalometer AE31) sampling inlet height was 3m and about 3m west of the closest highway lane. The two sites in student dormitories were located 60m and 1.4km from the highway and measured CO (Teledyne 300E), NOx (Thermo 42C), and BC (microAeth AE51). The site closer to the highway (Near Dorm or ND) operated out of an occupied administrative office on the ground floor of a five-story building and had an inlet height of 0.5m. The site further from the highway (Far Dorm or FD) operated out of an empty room part of a two bedroom-one bathroom suite on the ground floor of a five-story building and had an inlet height of 1.5m. Both dormitory sites included an automated valve on a 15-minute interval to alternate measurement of outdoor (NDO and FDO) and indoor (NDI and FDI) concentrations. The indoor inlet for both rooms was 0.25m off the flooring. The urban background (UB) site was located 2.3km west of the highway and was part of

the Southern Aerosol Research and Characterization (SEARCH) network. Previous studies have assessed this site as representative of Atlanta urban background pollutant concentrations and composition (Edgerton et al., 2005; Liu et al., 2005; Solomon et al., 2003).

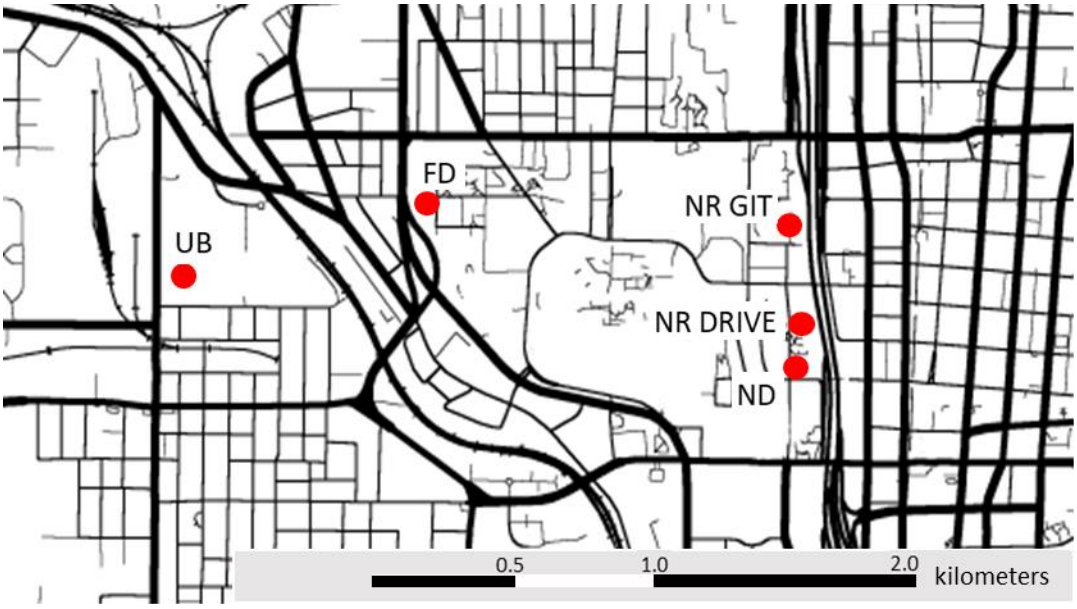

Figure 4.1 Sampling Map. NR GIT: Near-road Monitoring Network monitor on the GIT campus; NR-DRIVE: Near-road DRIVE site; ND: Near highway dorm outdoor and indoor sampling; FD: Far dorm outdoor and indoor sampling; UB: Urban background Jefferson St SEARCH site

Air quality monitors at the three DRIVE sites collected continuous ambient measurements from September 8, 2014 to January 5, 2015. Gas analyzers collected measurements at 5-second averaging periods using DAQFactory and WinWedge Pro

software. Multipoint calibrations with zero air and span checks provided an assessment for accuracy throughout the study and a time-weighted calibration was applied to the data based on calibrations. Particulate analyzers at the NR DRIVE, NR GIT, and UB sites collected BC measurements at 2-minute averaging periods. All continuous data were averaged to hourly levels to assess temporal variability differences between pollutants. The three DRIVE sites also collected 48-hr $PM_{2.5}$ measurements on teflon filters for the oxidative potential assay. All species and parameters were measured in local standard time (LST); however, daylight savings time did end on November 2, 2014. Traditional single pollutants were measured to generate and compare with multipollutant traffic indicators.

Part of the EPA Near-road Monitoring Network, Atlanta has two sites located along major highways. The first site (NR GIT) is located on the GIT campus adjacent to I-75/I-85 about 300m north of the NR DRIVE site. Trees were removed from a vegetation barrier along the highway to provide space for the site. Directly west of the site, there is a small limited-access parking lot for about 100 passenger vehicles. Monitoring for CO and NOx began on July 1, 2014 and for BC on November 3, 2014. The second EPA Near-road Monitoring Network site (NR SDK) began operation January 1, 2015 monitoring NOx and BC and is located 14km southeast of the NR GIT site in an open field adjacent to the interstate circling Atlanta (I-285). Both sites are operated by the Georgia Department of Natural Resources (GA EPD) with in inlet height of 3m and hourly concentration data were downloaded from the GA EPD air quality system (EPD).

4.2.2 Data Analysis

The Integrated Mobile Source Indicator (IMSI) was originally developed and evaluated using air quality concentration and mobile emissions data from Atlanta, Georgia to construct integrated estimates of vehicle emissions impact (Pachon et al., 2012), with a particular focus on its use for acute health impact analysis. By utilizing multiple single pollutant measurements, the metric provides a more stable value for characterizing exposure from both gasoline and diesel vehicle tailpipe emissions. The indicator values are derived from elemental carbon (EC), carbon monoxide (CO), and nitrogen oxide (NOx) concentrations normalized by the standard deviation (σ) of the hourly pollutant concentrations observed during the sampling period in order to combine concentrations of different magnitudes (Eq. 4.1). The species emissions ratios from vehicles were calculated as the fraction of the specific species emissions from mobile sources to total species emissions estimated from the 4km grid cell that includes all the sampling locations using the Sparse Matrix Operator Kernel Emissions (SMOKE) modelling system (CMAS), which uses the Mobile (MOVES) (EPA, 2010b).

(Eq. 4.1)

$$IMSI = \frac{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{Emis}\frac{EC}{\sigma_{EC}} + \left(\frac{NOx_{mob}}{NOx_{tot}}\right)_{Emis}\frac{NOx}{\sigma_{NOx}} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{Emis}\frac{CO}{\sigma_{CO}}}{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{Emis} + \left(\frac{NOx_{mob}}{NOx_{tot}}\right)_{Emis} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{Emis}}$$

where the measured concentrations are normalized by the standard deviation (σ) of the pollutant concentration. The emissions estimate ratios are calculated as the fraction of species emissions from mobile sources to the total species emissions. The source emissions were developed using the Sparse Matrix Operator Kernel Emissions (SMOKE) Modelling

System and are from the 4 km grid cell that includes the campus and the UB sampling site. Detailed information on the assumptions, mathematical derivation, and previous use of the metric is published elsewhere (Pachon et al., 2012).

4.2.3 Multivariate regression modeling

Data-driven regression modeling is a more readily applied method compared to the use of chemical transport models or dispersion models. The multivariate regressions provide a direct relationship for health studies to help understand and quantify the factors that drive near-road exposures for use in health analyses. To assess the factors that affected the temporal variability in the concentration of each TRAP as well as the IMSI, this study used a multivariate, linear, mixed regression model:

$$P_t = \beta\, Z_t + \theta_t + \varepsilon_t \qquad \text{(Eq. 4.2)}$$

where P_t denotes the concentration of BC, CO, NO, NO_2, NO_x, O_3, or IMSI measured during hour t and β is the coefficient of interest that describes the influence of factor Z_t on the hourly pollutant level. The factors assessed included time period of the day, temperature, wind speed, relative humidity, wind direction, weekend (Saturday and Sunday), and hourly traffic counts. The temporal factor was divided into five periods: morning rush hour (6 – 9am), mid-day (10am – 3pm), evening rush hour (4 – 8 pm), late evening (9pm – 12am), and early morning (1 – 5am). The wind direction factor was divided into three directions: north (315 – 45 degrees), east (45 – 135 degrees, which leads to the monitoring sites being downwind of the highway), and south (135 – 225 degrees). θ_t

represents time-specific random intercepts used to capture potential variations not explained by Z_t and ε_t represents residual random normal error. The regression relationship between pollutant concentrations or the multipollutant metric and driving factors were generated with R (version 3.5.0).

4.2.4 Biologically-relevant metric

The acellular assay measures the oxidative activity of water-soluble particles based on their ability to catalyze the transfer of electrons from dithiothreitol (DTT) to dissolved oxygen, generating superoxide radical anions. The rate at which DTT is depleted and ROS is generated mimics the biological process where DTT represents physiological antioxidants and oxidative stress is generated. When excess DTT is present in the system, the DTT consumption rate is proportional to the concentration of redox-active species in the $PM_{2.5}$ filter extracts. A variety of PM2.5 components emitted by vehicles, such as BC, polycyclic aromatic hydrocarbons (PAHs), oxygenated PAHs such as quinones and hydroxyquinones, and transition metal species have been linked to ROS generation (Cheung et al., 2010; de Kok et al., 2005; Surawski et al., 2010). The oxidative potential (OP) of atmospheric particulate samples is normalized by the volume of sampled air leading to units of nmol min^{-1} m^{-3} to provide the concentration of atmospheric aerosol OP. Alternatively, the OP of atmospheric particulate samples is normalized by the mass of the PM2.5 sample collected to provide the intrinsic atmospheric aerosol OP. The intrinsic OP is an indication of the toxicity of the measured aerosol and provides insight on the source specific contribution to aerosol OP.

4.3. Results and Discussion

To assess the variability and levels of single pollutant concentrations in comparison to the multipollutant traffic exposure indicator, hourly data for CO, NOx, and BC from the six sampling locations (the near-road (NR DRIVE) site, the Near-Road Network site (NR GIT), the near-highway dormitory outdoor (NDO) and indoor (NDI) sites, the far dormitory outdoor (FDO) and indoor (FDI) sites, and the urban background (UB) site) were compared to the hourly IMSI values at each site. Descriptive statistics and inter-site correlation analyses provided metrics to compare the temporal variability. Due to high concentrations that skewed the distribution causing a non-normal distribution, sites were compared using a Spearman's rank correlation. A more complete description of all the pollutants and meteorological parameters measured and analyzed, including personal exposure and biomarkers, are founds elsewhere (J. A. Sarnat et al., 2017).

4.3.1 Observed air pollutant and multipollutant metric dynamics

The IMSI show similar spatial trends as ambient CO, NOx, and BC concentrations with decreasing levels as the distance from the highway increases (Figure 4.2). The steepest portion of the gradient occurred within 60m from the highway between the NR DRIVE and NDO sites with a mean difference of 81 ppb (19%) for CO, 11 ppb (22%) for NOx, and 0.7 ug m^{-3} (41%) for BC. From the NR DRIVE to the FDO site (1.4km), the mean difference was 140 ppb (52%) for CO, 6 ppb (34%) for NOx, and 0.14 ug m^{-3} (50%) for BC. The IMSI follows a similar trend with a 14% decrease in value at the NDO site and a 40% decrease at the FDO site relative to the NR DRIVE site (Figure 4.3).

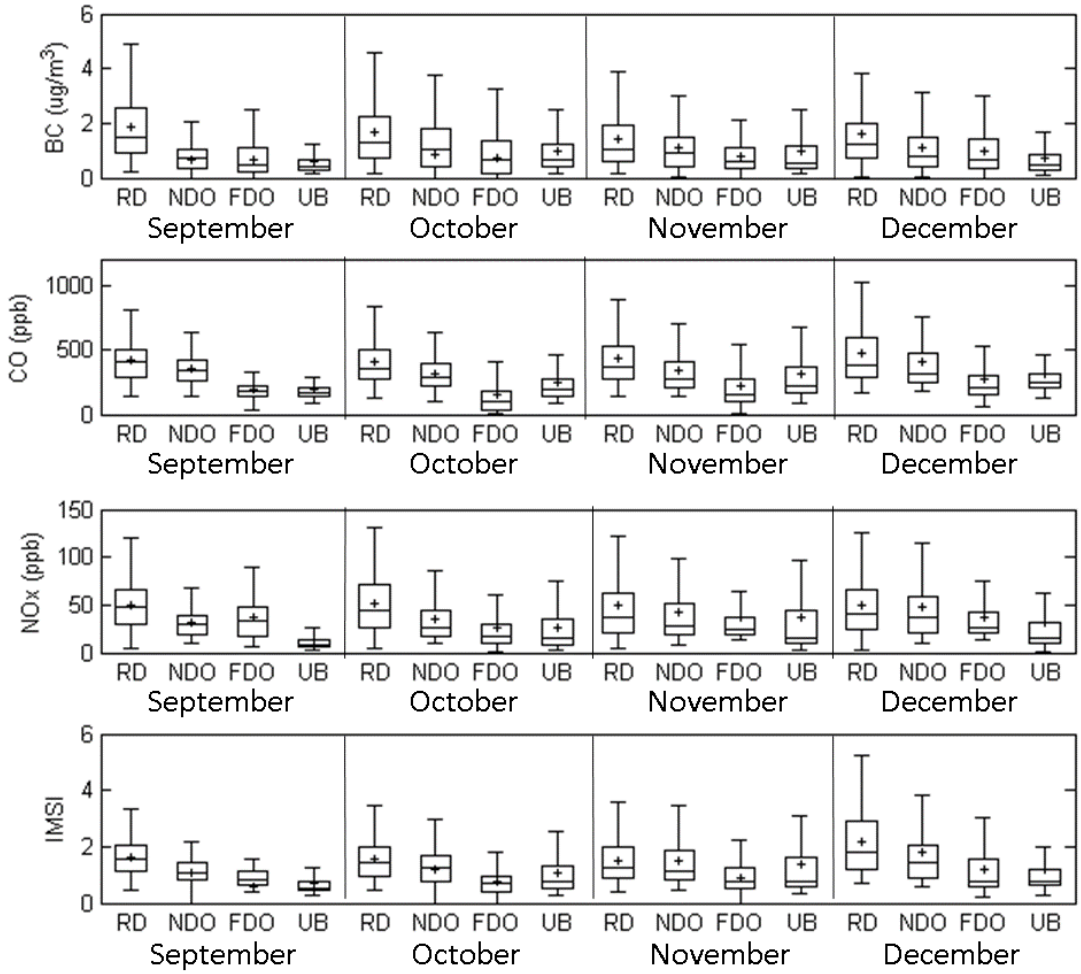

Figure 4.2 Boxplots of the hourly BC, CO, NOx, and IMSI levels from September 8th, 2014 to January 5th, 2015 at outdoor and indoor sampling locations, ordered in increasing distance from the highway source. RD - NR DRIVE (3m), NDO - Near Dorm outside (60m), FDO - Far Dorm outside (1.4 km), UB - Urban background (2.3 km)

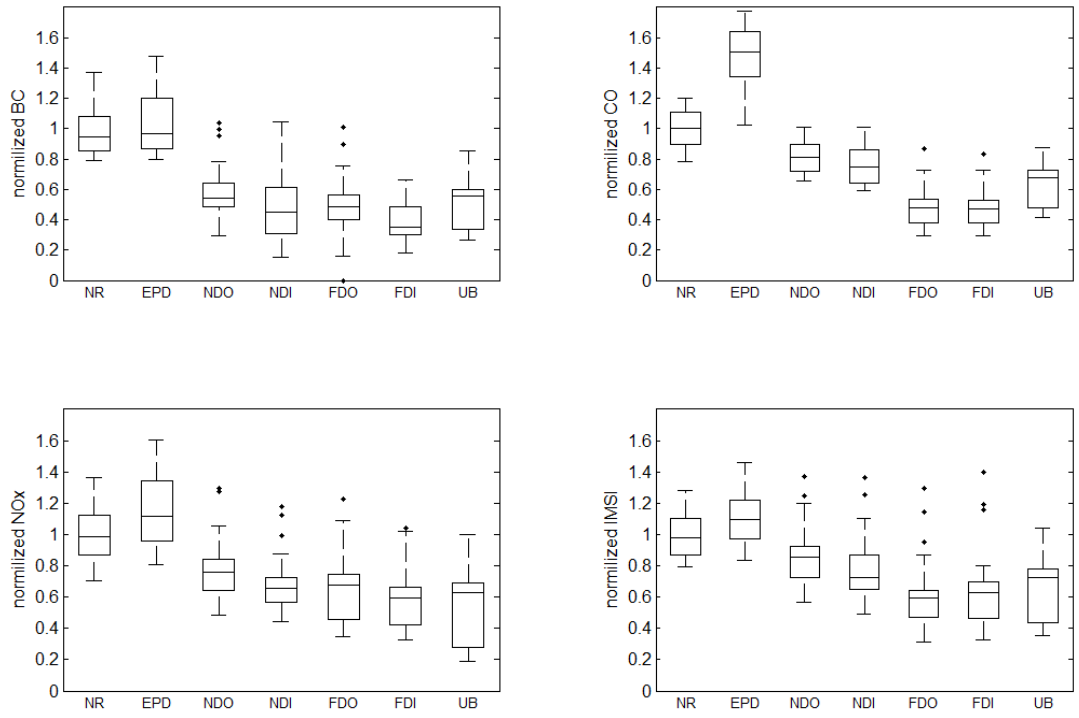

Figure 4.3 Normalized boxplot presenting the distribution of hourly BC, CO, NOx, and IMSI from Sept 8, 2014 to Jan 5, 2015, ordered in increasing distance from the highway source. NR – DRIVE Near-road (3m), EPD – NR GIT (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

The IMSI diurnal profile follows the patterns observed for the other primary pollutants with a peak in the morning as traffic increases and a decrease mid-day with an increase in the mixing height. The diurnal profile suggests chemical processing and transport influences the levels throughout the day leading to hours from 9am to 12pm with lower metric values near the highway (Figure 4.4). The mean normalized diurnal profiles for BC, CO, NOx, as well as the IMSI show the degree of daily variation for each pollutant

at the six sites (Figure 4.5). The normalized CO, NOx, and BC concentrations have similar diurnal profiles compared to the IMSI with a morning concentration peak at 9am, an evening peak at 9pm, and minimum concentrations observed at 3am and 4pm. With increasing distance from the highway, the diurnal variability of the normalized concentrations of the primary species increase from about 0.8 to 1.5 times the mean NR DRIVE concentration compared to about 0.5 to 2.2 times the mean FDO concentration for all species and the multipollutant metric. With increasing distance from the major source of vehicle emissions, the diurnal variability becomes more drastic for CO, NOx, BC, and the IMSI. This is further reflected in the concentration difference between the diurnal maximum and minimum concentrations. At the NR DRIVE site, the CO, NOx, BC, and IMSI difference in the diurnal maximum and minimum is 178ppb, 33ppb, 0.93 ug m^{-3}, and 0.8 compared to the 245ppb, 44ppb, 1.62 ug m^{-3}, and 1.6 difference at the FDO site, respectively. While the mixing height varies enough throughout the day to decrease the concentration at the NR site, direct vehicle emissions associated with the consistently high daytime traffic count throughout the day from 7 am to 7 pm maintains a minimum concentration of about 0.8 times the mean concentration at the NR DRIVE site. Chemical processing and transport leads to lower normalized minimum concentrations and higher normalized maximum concentrations at the FDO site.

Similar to the outdoor concentrations, the indoor pollutant observations peaked in the morning and evening. For BC and CO, the outdoor and indoor peaks occurred at the same time, but for NOx the morning peak observed an hour lag in the observed maximum indoor morning concentration. Concentrations were also similar suggesting the high infiltration rate for both the gaseous and aerosol species.

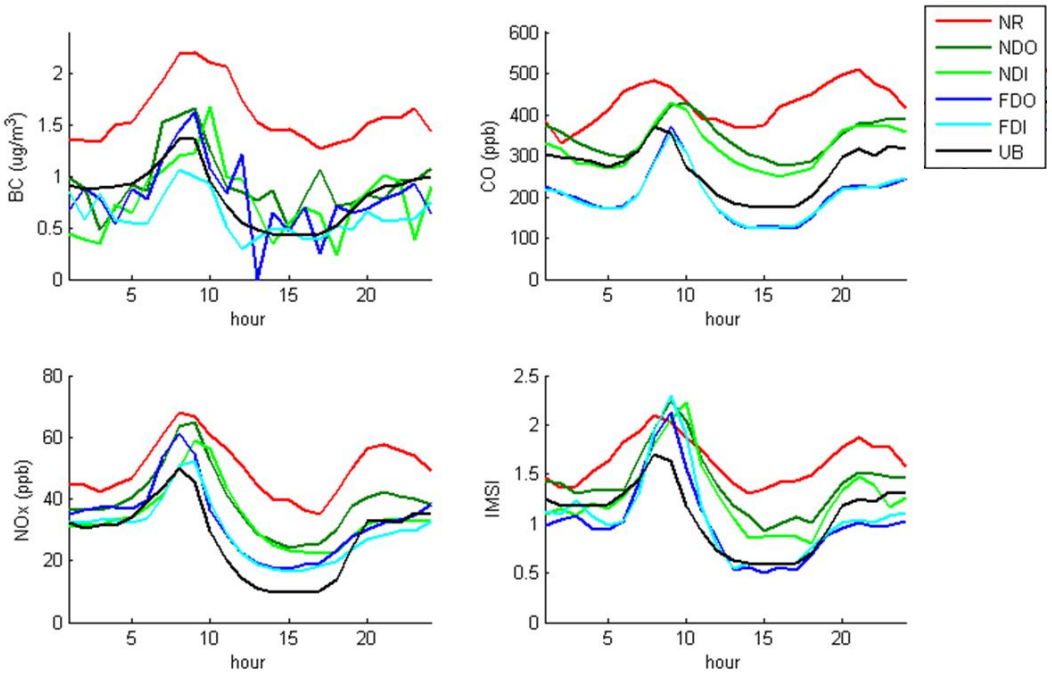

Figure 4.4 Diurnal profiles for hourly concentrations of BC, CO, NOx, and IMSI for the DRIVE sites from September 8, 2014 to January 5, 2015. NR – DRIVE Near-road (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

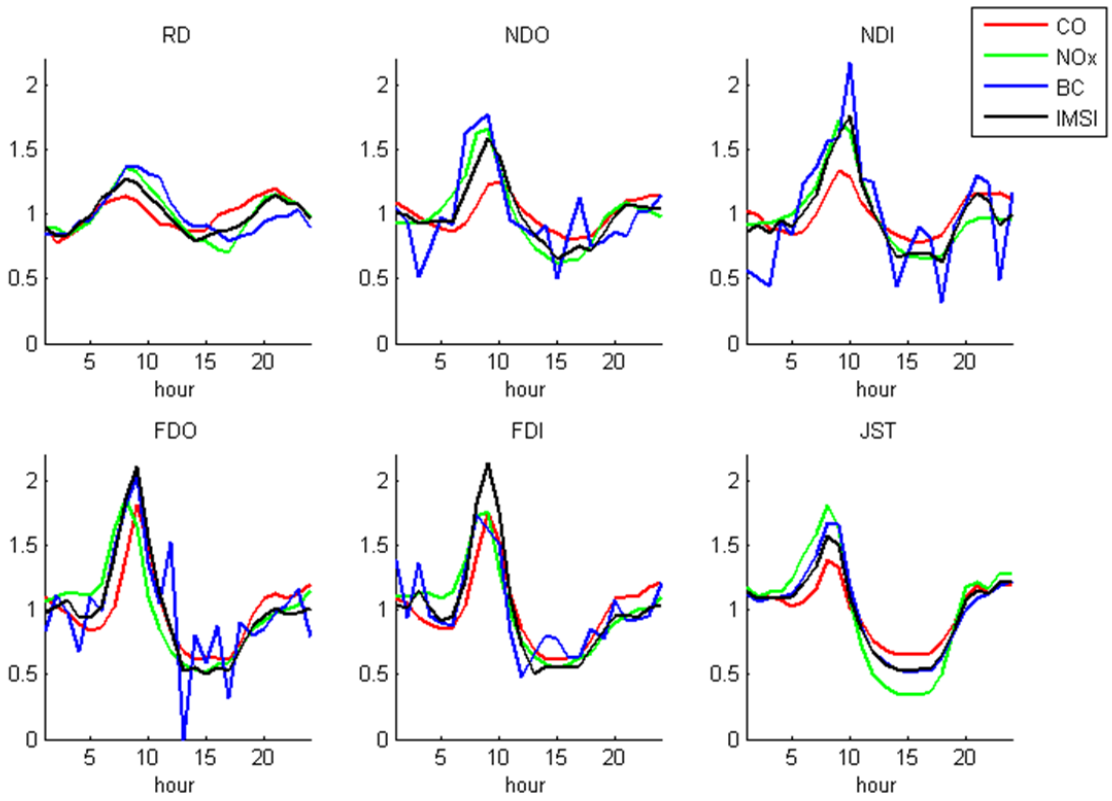

Figure 4.5 Normalized concentration from Sept 8, 2014 to Jan 5, 2015, ordered in increasing distance from the highway source. RD – NR DRIVE (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

4.3.2 Spatial and temporal correlations

Site correlations examine how well hourly temporal variability patterns between the six monitoring locations reflected corresponding temporal variability at the other sites with varying distances from each other based on all available data during the sampling period (Table 4.1). With increasing distance, the IMSI Spearman's correlation between the NR DRIVE site and the other outdoor sites decreases. The individual pollutants however do not observe this correlation trend. The BC concentrations at the NR GIT and UB sites

are more correlated with the NR DRIVE site than the NDO site. The NOx and CO concentrations at the NDO site however were more correlated with the NR DRIVE site than the NR GIT site. Site specific properties likely impact the concentration spatial gradient depending on the pollutant. NO_2 concentrations measured at the NR DRIVE were more temporally correlated across the domain than the three other primary traffic species (BC, CO, and NO) with observed Spearman's correlations greater than 0.7 between the NR DRIVE site and the other ambient sites (data not shown). For BC, CO, and NO, stronger correlations with the NR DRIVE site were generally found for sites closer to the NR DRIVE site compared to those further away.

The correlation between the ND and the FD sites were higher for the gaseous NOx (0.67) and CO (0.70) pollutants than the BC (0.46) concentration. These correlation trends are consistent with the spatial gradients observed for the species. The gaseous pollutants have more homogenous concentrations and regional sources. CO is emitted as a primary pollutant from vehicle and generated as a secondary pollutant from VOC oxidation. Similarly, NOx is both emitted as a primary pollutant and generated as a secondary pollutant as a result of long-range transport. These additional sources as well as lower deposition rates lead to higher correlations among the sites. Black carbon is a primary pollutant with a lower lifetime in the atmosphere leading to a greater spatial gradient and a lower correlation between the two dormitory monitoring locations. By using the IMSI as a metric for exposure, the Spearman's correlation (0.72) between the two locations was greater than the correlation for any of the individual species.

In addition to difference in correlations between the buildings, the infiltration rate of the pollutants varied based on species and building. For the ND site, the correlation

between the ambient and outdoor measurements were higher for NOx (0.93) and CO (0.90) compared to BC (0.53). This trend was also observed at the FD site where the correlation was 0.96 for NOx and 0.97 for CO compared to 0.51 for BC. Building filtration systems are designed to capture particles, though even the gaseous pollutants were captured at a higher efficiency with the newer system installed at the ND building. Compared to the species, the IMSI metric shows a correlation between the ambient and indoor environments of 0.80 at the ND and 0.87 at the FD.

Table 4. 1 Spearman's Correlation for BC, NOx, CO, and IMSI between sampling locations for hourly levels from September 8, 2014 to January 5, 2015

BC \ NOx	NR DRIVE	NR GIT	NDO	NDI	FDO	FDI	UB	Average
NR DRIVE		0.71	0.85	0.80	0.61	0.63	0.64	0.71
NR GIT	0.80		0.70	0.70	0.58	0.58	0.49	0.63
NDO	0.56	0.78		0.93	0.67	0.67	0.64	0.74
NDI	0.52	0.75	0.53		0.64	0.68	0.62	0.73
FDO	0.39	0.55	0.46	0.35		0.96	0.78	0.71
FDI	0.34	0.47	0.42	0.34	0.51		0.78	0.72
UB	0.64	0.69	0.59	0.50	0.50	0.51		0.66
Average	0.54	0.67	0.56	0.50	0.46	0.43	0.57	0.53 \ 0.70

CO \ IMSI	NR DRIVE	NR GIT	NDO	NDI	FDO	FDI	UB	Average
NR DRIVE		0.78	0.74	0.69	0.68	0.63	0.65	0.70
NR GIT	0.44		0.81	0.81	0.72	0.65	0.67	0.74
NDO	0.66	0.52		0.80	0.72	0.70	0.65	0.74
NDI	0.67	0.53	0.90		0.68	0.66	0.62	0.71
FDO	0.50	0.48	0.70	0.68		0.87	0.78	0.74
FDI	0.52	0.49	0.72	0.70	0.97		0.76	0.71
UB	0.48	0.34	0.55	0.58	0.74	0.77		0.69
Average	0.55	0.47	0.68	0.68	0.68	0.70	0.58	0.62 \ 0.72

While the two near-road sites are located along the same highway segment, the NR DRIVE site was located in an open parking lot and the NR GIT site is in line with a vegetation barrier. A vegetation barrier impacts the rate is dispersion leading to different pollutant dynamics even along the same roadway segment (R. Baldauf, 2017; Janhall, 2015). By including three TRAPs in a single multi-pollutant value, the IMSI is not as impacted by differences in site location properties.

Diurnal site correlations further examine how the temporal variability of the vehicle emissions and meteorological conditions affect the concentrations measured at sites with increasing distance from the highway. The diurnal correlation between the measurements at the NR DRIVE site and the other sites showed the correlation was strongest in the morning consistent with the idea that the highway emissions are a major local source effecting concentrations (Figure 4.6). Further, as traffic increases in the morning, vertical diffusion is limited resulting in higher, more consistent concentration levels across sites.

Decreased correlation is observed throughout the day as both photochemical reactions and vertical mixing increases. A decrease in correlation between sites for CO occurs earlier in the day than for NOx. The CO correlations began to decrease at 5am, reached a minimum correlation at 1pm, and continued to improve throughout the afternoon. While the CO correlation with the NDO site was between 0.49 and 0.82, the correlation with the FDO site was much wider between 0.12 and 0.74. The NOx correlations began to decrease later in the day at 10am and remained at a minimum from 2pm to 7pm. The correlations with the NDO site were between 0.74 and 0.91, and the correlations with the FDO site were between 0.23 and 0.81. For CO and NOx, the correlations between the NR DRIVE and NDO sites were consistently better than the correlations with the FDO site for

any given hour suggesting the correlation decreased with distance. However, the correlations with the NR DRIVE site and the UB site varied such that they were better than the NDO or FDO sites at certain hours. Due to the complexity of how single pollutants disperse from major highway sources, near-road monitors lead to exposure measurement errors (as compared to instrument measurement errors) when used to determine exposures for individuals at the far dorm (D. H. Liang et al., 2018). While the correlations between the NR DRIVE site and the other sites still decrease throughout the day, it is more consistent across the sites. The minimum diurnal IMSI correlation between the NR DRIVE site and the NDO site was 0.50 and for the UB site, it was 0.38.

The comparison of the range of the diurnal variability of the IMSI and the individual species correlations between the NR DRIVE site and the other monitoring site locations shows the ability of the IMSI to improve the spatial correlations. The trend of the diurnal Spearman's correlations between the IMSI and the species for the correlations between the NR GIT and NR DIRVE sites were relatively flat with a wide range of correlations observed in the IMSI value and a smaller range observed in the single species. With increasing distance between the correlation sites, the range in the correlation for the ND site and the NR DIRVE site for the IMSI values shrunk leading to a cluster of points with higher correlations. The trend of the correlations between the FD site and the NR DRIVE site were scattered with a trend around the one-to-one line. While the UB site was the furthest site from the NR DRIVE site, the correlation of the site correlations was less scattered than observed at the FD sites.

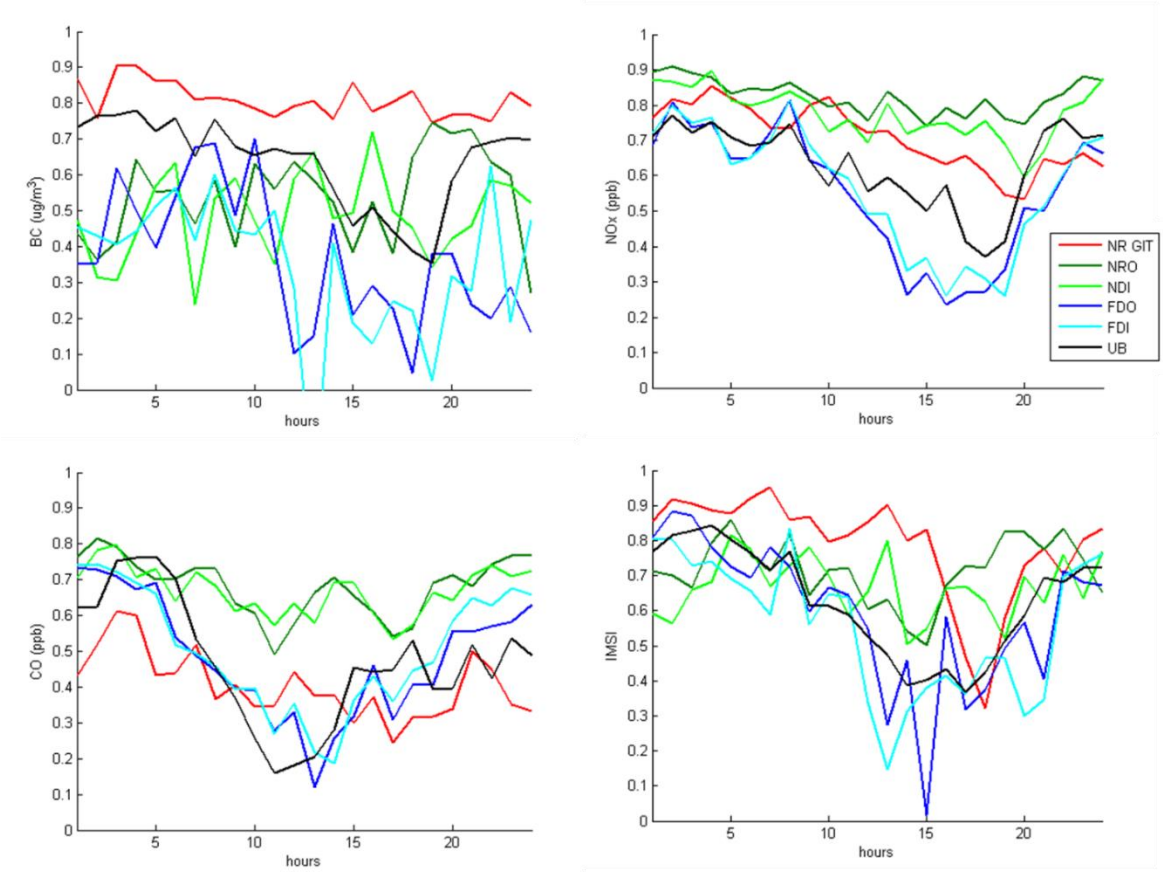

Figure 4.6 Diurnal profile of Spearman's correlation between the NR DRIVE site and the other sites from September 8, 2014 to January 5, 2015. NR GIT – EPA Near-road Monitoring (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

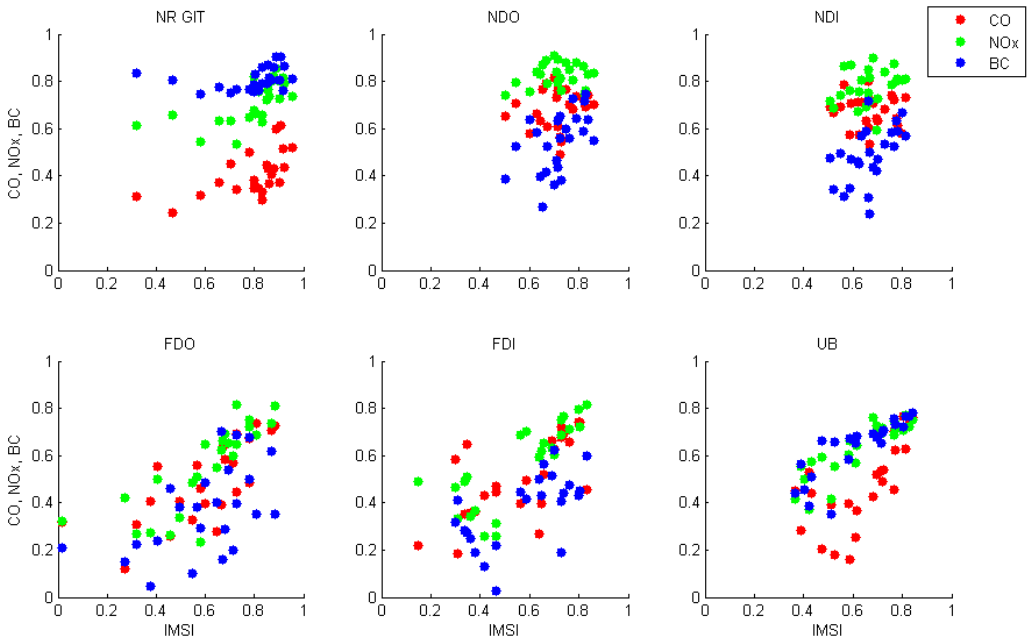

Figure 4.7 Regression of diurnal Spearman's correlation between the NR DRIVE site and the other sites for the single species and the IMSI values. NR GIT – EPA Near-road Monitoring (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

4.3.3 Assessment of factors driving metric variability

Since the NAAQS for both NO_2 and CO include primary standards based on an hourly concentration, the DRIVE study aimed at understanding exposures to individuals in the near-road environment with hourly concentrations. The meteorological and traffic conditions were also assessed hourly so that these factors could be linked to the hourly concentrations. Statistical modeling can help identify the significant factors that affect the observations at different monitoring locations. Here, linear mixed modeling evaluated associations between pollutant concentrations and multiple possible contributing predictors

was used to assess factors that drive the temporal variability observed at the different near-road sites. The regression coefficients for the models developed for the NR DRIVE site and the NR GIT site were compared to assess whether site differences along the same road segment can lead to significant differences in the dominate factors driving primary pollutants or multipollutant metric (Table 4.2). The regression coefficients for two sites at varying distances from the highway (NR GIT and UB) were also compared to assess how exposure analyses may vary based on proximity to roadways in an urban area (Table 4.3). Significance of a factor was determined by a p-value less than 0.05.

During the DRIVE study period, the NR DRIVE and NR GIT site regression coefficients for BC, CO, and NOx concentrations as well as the IMSI metric were negatively associated with wind speed and wind direction from the north, east, and south indicative of dispersion away from the emissions source (Table 4.2). Temperature was only associated with a decrease in concentration at the NR GIT site and was associated with an increase in BC and CO concentration at the NR DRIVE site. While mixing height was associated with a significant decrease in BC and NOx concentration at the NR DRIVE site and in CO and NOx concentration at the NR GIT site, mixing height did not have a significant coefficient at either site for the IMSI metric suggesting the IMSI is a more stable metric for exposure. Weekend days showed an association with a significant decreasing concentration for all pollutants (NO, NO_2, and BC) except CO at both sites. For the IMSI metric, weekends were significant at the NR GIT site but not at the NR DRIVE site. Traffic count was associated with a significant increase in concentration and multipollutant metric values at both sites, except for CO and the NR GIT site. Overall, the IMSI metric had fewer

significant terms associated with the factors assessed suggesting that the IMSI would be a more stable way to assess exposure compared to single pollutants.

Differences in the regression coefficients for the NR DRIVE and NR GIT sites are likely linked to the differences in the physical site locations. The NR DRIVE site is located in an open parking lot, while the NR GIT site was located in line with a vegetation barrier along the highway. Although the two sites are within 300m and along the same highway segment, the differences in their behavior has important implications for exposure analyses when relying on a single central monitor.

Table 4.2 Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations from September 8, 2014 to January 5, 2015. *All covariates were included simultaneously in the model for each pollutant of interest

NR DRIVE	Black Carbon		Carbon Monoxide		Nitrogen Oxides		IMSI	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-12am)	0.23*	(0.04,0.42)	6.08	(-27.25,39.41)	5.00*	(0.35,9.65)	0.13*	(0.01,0.26)
Early Morning (1-5am)	0.24*	(0.03,0.44)	-8.52	(-44.92,27.88)	3.18	(-1.82,8.18)	0.10	(-0.03,0.24)
Morning Rush Hour (6-9am)	0.39*	(0.13,0.64)	-11.98	(-55.88,31.92)	5.65	(-0.34,11.64)	0.13	(-0.04,0.29)
Mid Day (10am-3pm)	0.14	(-0.04,0.32)	-27.31	(-58.23,3.61)	1.28	(-3.00,5.57)	-0.01	(-0.13,0.10)
Temperature	0.01*	(0.00,0.02)	2.35*	(0.74,3.97)	-0.06	(-0.28,0.16)	0.00	(-0.01,0.01)
Relative Humidity	0.00	(0.00,0.01)	-0.37	(-1.31,0.58)	0.06	(-0.07,0.20)	0.00	(0.00,0.00)
Wind Speed	-0.20*	(-0.26,-0.14)	-40.75*	(-50.09,-31.40)	-6.88*	(-8.19,-5.56)	-0.21*	(-0.25,-0.18)
Northerly Wind	-0.27*	(-0.47,-0.08)	-36.78*	(-70.63,-2.92)	-10.04*	(-14.88,-5.20)	-0.21*	(-0.33,-0.10)
Easterly Wind	-0.73*	(-0.95,-0.51)	-59.43*	(-97.25,-21.61)	-29.25*	(-34.65,-23.85)	-0.47*	(-0.60,-0.34)
Southerly Wind	-0.30	(-0.76,0.17)	-7.17	(-83.98,69.64)	-23.95*	(-35.24,-12.65)	-0.28*	(-0.55,0.00)
Weekend	-0.61*	(-0.97,-0.26)	-11.84	(-74.45,50.77)	-14.03*	(-21.95,-6.10)	-0.28	(-0.56,0.00)
Traffic Counts (per 1,000)	0.06*	(0.04,0.07)	8.60*	(6.56,10.64)	1.58*	(1.30,1.86)	0.04*	(0.03,0.05)
Mixing Heights (per 100 meters)	-0.03*	(-0.05,-0.01)	-3.41	(-7.12,0.30)	-0.99*	(-1.53,-0.45)	-0.01	(-0.03,0.00)

Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for Black Carbon: µg/m3; Unit for Carbon Monoxide, Nitrogen Oxides: ppb

NR GIT	Black Carbon		Carbon Monoxide		Nitrogen Oxides		IMSI	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-12am)	0.54*	(0.29,0.79)	47.63*	(6.97,88.29)	11.39*	(6.59,16.19)	0.35*	(0.15,0.56)
Early Morning (1-5am)	0.52*	(0.24,0.79)	-22.45	(-66.23,21.34)	10.85*	(5.58,16.13)	0.25*	(0.03, 0.48)
Morning Rush Hour (6-9am)	0.70*	(0.37,1.03)	-1.59	(-54.11,50.94)	14.18*	(7.94,20.41)	0.32*	(0.05, 0.59)
Mid Day (10am-3pm)	0.49*	(0.25,0.72)	25.87	(-11.83,63.57)	7.94*	(3.50,12.37)	0.24*	(0.05, 0.43)
Temperature	-0.02*	(-0.04,-0.00)	-4.00*	(-6.13,-1.86)	-0.89*	(-1.11,-0.66)	-0.03*	(-0.04, -0.01)
Relative Humidity	0.00	(-0.01,0.01)	-0.33	(-1.59,0.93)	0.00	(-0.14,0.13)	-0.01	(-0.01, 0.00)
Wind Speed	-0.19*	(-0.27,-0.12)	-45.35*	(-57.35,-33.34)	-5.33*	(-6.70,-3.96)	-0.20*	(-0.26, -0.14)
Northerly Wind	-0.74*	(-1.28,-0.20)	-90.25*	(-134.28,-46.23)	-12.45*	(-17.54,-7.36)	-0.77*	(-1.20, -0.34)
Easterly Wind	-1.25*	(-1.79,-0.70)	-191.16*	(-240.46,-141.87)	-25.53*	(-31.20,-19.85)	-1.23*	(-1.66, -0.79)
Southerly Wind	-	-	-222.11*	(-324.75,-119.47)	-29.55*	(-41.41,-17.70)	-	-
Weekend	-0.81*	(-1.40,-0.22)	-9.13	(-92.74,74.49)	-15.40*	(-24.01,-6.80)	-0.46*	(-0.89, -0.04)
Traffic Counts (per 1,000)	0.75*	(0.60, 0.90)	216.00	(191.11, 240.89)	27.60*	(24.68, 30.52)	0.64*	(0.51, 0.77)
Mixing Heights (per 100 meters)	-0.09	(-0.42, 0.24)	-63.9*	(-111.45, -16.35)	-6.36*	(-11.89, -0.83)	-0.17	(-0.44, 0.10)

Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for Black Carbon: µg/m3; Unit for Carbon Monoxide, Nitrogen Oxides: ppb

Table 4.2 (continued)

UB	Black Carbon		Carbon Monoxide		Nitrogen Oxides		IMSI	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-12am)	1.00*	(0.03, 0.22)	20.7	(-3.1, 44.5)	3.20	(-1.1, 7.5)	0.12*	(0.01, 0.23)
Early Morning (1-5am)	0.65	(-0.04, 0.16)	-11.7	(-37.2, 13.7)	-0.04	(-4.7, 4.7)	0.00	(-0.12, 0.12)
Morning Rush Hour (6-9am)	0.10	(-0.03, 0.22)	-2.9	(-33.8, 28.1)	0.24	(-5.5, 6.0)	0.03	(-0.12, 0.19)
Mid Day (10am-3pm)	0.00	(-0.08, 0.09)	-7.0	(-28.9, 15.0)	-0.49	(-4.5, 3.5)	-0.02	(-0.12, 0.08)
Temperature	-0.01*	(-0.01, -0.00)	-6.3*	(-7.6, -5.9)	-0.99*	(-1.2, -0.8)	-0.03*	(-0.03, -0.02)
Relative Humidity	0.01*	(0.00, 0.01)	1.8*	(1.2, 2.5)	0.23*	(0.1, 0.4)	0.01*	(0.01, 0.01)
Wind Speed	-0.29*	(-0.33, -0.25)	-69.6*	(-79.7, -59.5)	-13.2*	(-15.0, -11.4)	-0.16*	(-0.40, -0.30)
Northerly Wind	0.06	(-0.00, 0.13)	41.3*	(24.4, 58.2)	8.59*	(5.6, 11.6)	0.17*	(0.09, 0.25)
Easterly Wind	0.01	(-0.08, 0.10)	9.4	(-13.3, 32.1)	5.67*	(1.6, 9.8)	0.05	(-0.06, 0.15)
Southerly Wind	-0.05	(-0.12, 0.03)	-20.4*	(-39.9, -1.0)	-0.87	(-4.3, 2.6)	-0.07	(-0.16, 0.02)
Weekend	-0.15	(-0.40, 0.06)	1.1	(-56.2, 58.3)	-6.11	(-15.5, 3.3)	-0.12	(-0.40, 0.15)
Traffic Counts (per 1,000)	0.02*	(0.01, 0.02)	3.8*	(2.4, 5.2)	0.59*	(3.2, 8.6)	0.02*	(0.01, 0.02)
Mixing Heights (per 100 meters)	-0.01	(-0.01, 0.01)	2.8	(-0.04, 5.6)	0.22	(-0.3, 0.7)	0.01	(-0.00, 0.02)

Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for Black Carbon: µg/m3; Unit for Carbon Monoxide, Nitrogen Oxides

Table 4.3 Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations from Jan 1, 2015 to Dec 31, 2015. *All covariates were included simultaneously in the model for each pollutant of interest

NR GIT	Black Carbon		Carbon Monoxide		Nitrogen Oxides		IMSI	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-12am)	0.85*	(0.60, 1.09)	94.5*	(50.5, 138.4)	13.2*	(8.5, 18.0)	0.39*	(0.26, 0.51)
Early Morning (1-5am)	0.18*	(0.91, 1.44)	87.3*	(40.9, 133.7)	18.9*	(13.8, 24.0)	0.54*	(0.40, 0.68)
Morning Rush Hour (6-9am)	0.68*	(1.37, 2.00)	113.4*	(59.1, 167.7)	26.0*	(20.0, 32.0)	0.70*	(0.54, 0.87)
Mid Day (10am-3pm)	0.57*	(0.33, 0.81)	2.5	(-40.0, 45.0)	6.5*	(1.9, 11.0)	0.18*	(0.05, 0.30)
Temperature	-0.01	(-0.02, 0.00)	-9.6*	(-11.9, -7.3)	-0.9*	(-1.1, -0.6)	-0.02*	(-0.03, -0.01)
Relative Humidity	-0.00	(-0.00, 0.00)	0.5	(-0.2, 1.2)	-0.1	(-0.1, 0.02)	0.00	(-0.00, 0.00)
Wind Speed	0.19*	(0.8, 0.30)	34.1*	(15.3, 52.9)	7.0*	(5.1, 9.0)	0.17*	(0.11, 0.22)
Northerly Wind	-0.54*	(-0.95, -0.13)	-136.2*	(-208.0, -64.4)	-14.6*	(-21.6, -7.6)	-0.32*	(-0.54, -0.11)
Easterly Wind	0.18	(-0.12, 0.48)	65.5*	(9.1, 121.9)	7.4*	(1.9, 13.1)	0.16	(-0.00, 0.32)
Southerly Wind	0.22*	(0.01, 0.42)	41.5*	(4.3, 78.8)	4.8*	(1.1, 8.5)	0.11	(-0.00, 0.22)
Weekend	-1.69*	(-2.09, -1.29)	-173.5*	(-297.9, -49.2)	-28.1*	(-36.6, -19.5)	-0.73*	(-1.00, -0.46)
Traffic Counts (per 1,000)	0.11*	(0.09, 0.13)	32.0*	(29.1, 34.9)	2.6*	(0.0, 0.0)	0.08*	(0.07, 0.09)
Mixing Heights (per 100 meters)	-	-	-	-	-		-	-

Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for Black Carbon: µg/m3; Unit for Carbon Monoxide, Nitrogen Oxides

UB	Black Carbon		Carbon Monoxide		Nitrogen Oxides		IMSI	
	Est.	95% CI	Est.	95% CI	Est.	95% CI	Est.	95% CI
Late Evening (9pm-12am)	-0.00	(-0.11, 0.09)	-8.3	(-32.9, 16.3)	-0.88	(-4.2, 2.7)	-0.01	(-0.14, 0.11)
Early Morning (1-5am)	0.01	(-0.10, 0.12)	-56.0*	(-82.5, -29.5)	-2.0	(-5.7, 1.8)	-0.04*	(-0.28, -0.00)
Morning Rush Hour (6-9am)	-0.07	(-0.20, 0.06)	-61.6*	(-94.1, -29.1)	-5.9*	(-10.3, -1.4)	-0.24*	(-0.41, -0.07)
Mid Day (10am-3pm)	0.04	(-0.05, 0.13)	-4.1	(-26.5, 18.3)	-0.14	(-3.3, 3.0)	0.00	(-0.11. 0.12)
Temperature	-0.02*	(-0.03, -0.01)	-0.35	(-2.6, 1.9)	-0.53*	(-0.8, -0.2)	-0.01*	(-0.02, -0.00)
Relative Humidity	0.02*	(0.02, 0.02)	7.6*	(6.7, 8.6)	0.75*	(0.6, 0.9)	0.04*	(0.03, 0.04)
Wind Speed	-0.06*	(-0.09, -0.04)	-14.8*	(-20.4, -9.2)	-2.74*	(-3.5, -2.0)	-0.09*	(-0.12, -0.06)
Northerly Wind	-0.02	(-0.10, 0.06)	25.3*	(6.9, 43.6)	9.6*	(7.0, 12.2)	0.13*	(0.04, 0.23)
Easterly Wind	-0.12*	(-0.22, -0.02)	-14.3	(-38.8, 10.1)	0.97	(-2.5, 4.4)	-0.07*	(-0.19, 0.06)
Southerly Wind	-0.16*	(-0.24, -0.08)	-31.4*	(-50.8, -12.0)	-2.7	(-5.4, 0.08)	-0.18*	(-0.28, -0.08)
Weekend	-0.51*	(-0.87, -0.16)	-70.4	(-182.0, 41.3)	-11.1	(-23.4, 1.2)	-0.49	(-1.08, 0.10)
Traffic Counts (per 1,000)	0.02*	(0.02, 0.03)	4.4*	(2.9, 5.9)	0.71*	(0.5, 0.9)	0.03*	(0.02, 0.03)
Mixing Heights (per 100 meters)	-	-	-	-	-	-	-	-

Est. Estimate of Coefficient; 95% CI- 95% Confidence Interval; Unit for Black Carbon: µg/m3; Unit for Carbon Monoxide, Nitrogen Oxides

4.3.4. Biologically-relevant metric

The biologically-relevant metric utilized in this study was an acellular assay that quantifies oxidative potential of PM2.5 samples based on the consumption rate of DTT. Oxidative potential was assessed as a marker for toxicity of primary exhaust emissions and the important of atmospheric processing of PM for the potential tie to health outcomes from PM2.5 exposure. The spatial distribution of the oxidative potential of the water-soluble fraction was compared to the spatial gradients observed in the EC, OC, and PM2.5 mass integrated, filter measurements (Figure 4.8). The average EC concentration was 1.1 ug m^{-3} at the NR DRIVE site, 0.8 ug m^{-3} at the NDO site, and 1.0 ug m^{-3} at the FDO site. The OC mass measurements showed an increase with distance from 2 ug m^{-3} at the NR DRIVE site to 4 ug m^{-3} at the NDO site and 5 ug m^{-3} at the FDO site. Similarly, the PM2.5 mass showed an increase with distance from 7.5 ug m^{-3} at the NR DRIVE site to 12ug m^{-3} at the NDO site and 13 ug m^{-3} at the FDO site. The increasing trend in mass concentration is likely due to the increasing composition of secondary species in the particles as primary vehicle emissions react. The lack of elevated oxidative potential observed in the near-road environment compared to the other sites further from the highway suggests that atmospheric processing with the oxidation of primary PAHs to quinones and hydroxyquinones and metals further enhances oxidative potential of PM2.5 when components are more oxidized and water-soluble. A spatial gradient is observed in the near-road environment for total $PM_{2.5}$, which considered the insoluble fraction (Gao et al., 2017).

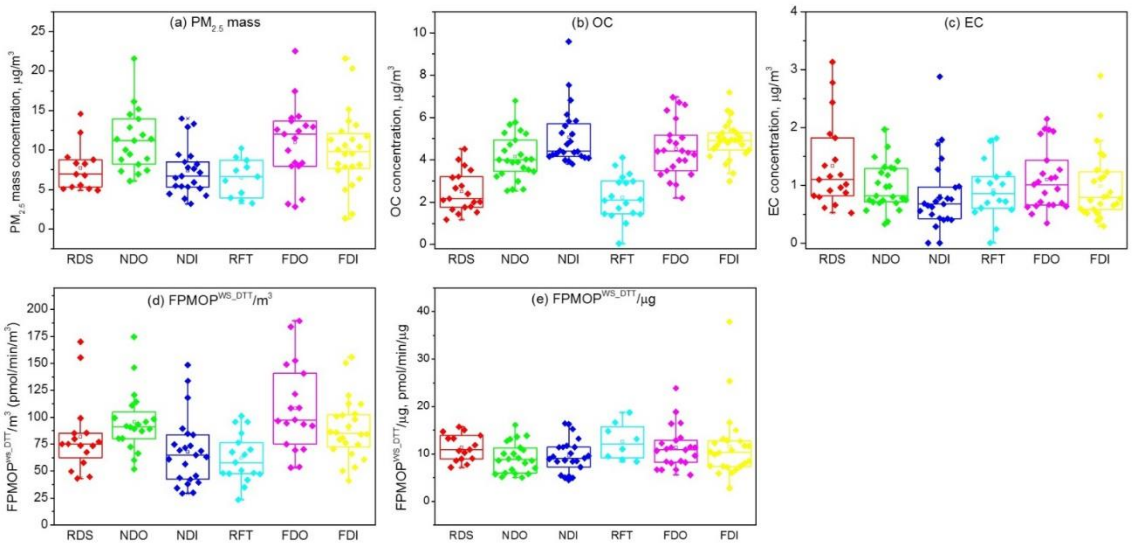

Figure 4.8 Spatial distribution of concentrations of $PM_{2.5}$ mass, OC, EC, volume normalized OP, and mass normalized OP from 48-hour integrated filter measurements at the sampling sites ordered in increasing distance from roadway.

Intrinsic oxidative potential is OP normalized by the PM2.5 mass and is an indication of the toxicity of the aerosols. The measurement of intrinsic OP also provides an insight on the sources linked to the overall OP. Previous source apportionment studies have found light-duty gasoline vehicles have high intrinsic OP, followed by biomass burning and heavy-duty diesel vehicle emissions (Bates et al., 2015). Since the mass and volume normalized oxidative potential showed little spatial gradient, primary vehicle emissions did not contribute substantially to the measured oxidative potential compared to the regional contribution. Source contribution can further be assessed through the correlation of oxidative potential to the single species measured. The mass and volume normalized oxidative potential was highly correlated with OC as well as EC, CO, and NOx, though the correlation with normalized OP was greater for the species with similar spatial

gradients. The regression analysis showed a high correlation ($R^2 > 0.6$) between volume normalized OP and OC at the NR DRIVE, ND, and FDO sites as well as EC and $PM_{2.5}$ mass at the FD sites.

The comparison of ambient and indoor correlations for the single species measured provides insights on outdoor infiltration and indoor course that are also contributing to the indoor measurements. At the two dormitory sites, the ambient and indoor mass normalized oxidative potential data were of similar magnitudes. At the ND site, however, PM2.5 mass, BC, and volume normalized OP were higher in the ambient measurements compared to the indoor measurements, with the opposite trend observed in the OC concentrations. The correlation between the ambient and indoor measurements for the single pollutants and oxidative potential were high ($r > 0.6$) for all species at the FD. For the ND site, the gas species (CO and NOx) were highly correlated ($r > 0.97$) and the aerosol components ($PM_{2.5}$ mass and OP) were not as well correlated ($r = 0.6$). These correlations suggest higher filtration rates for gas species as well as particulates at the FD site, which had an older HVAC system. These correlations also suggest that for OP there was no significant source within the dormitory rooms given the similar mass normalized OP measured both ambient and indoors.

Table 4.4 Correlation (Pearson's r) between volume normalized OP at the sampling locations with the single species pollutants

	NR DRIVE (N = 18)	NDO (N = 20)	NDI (N = 21)	FDO (N = 17)	FDI (N = 17)
$PM_{2.5}$ mass	**0.87**	0.58	**0.81**	**0.77**	0.68
OC	**0.88**	**0.91**	**0.86**	**0.90**	**0.80**
EC	**0.77**	0.69	**0.85**	**0.91**	**0.91**
CO	0.56*	**0.71**	0.60	**0.75**	0.59
NO_x	0.46*	**0.79**	**0.76**	**0.76**	**0.76**
BC	0.60	0.56	**0.75**	**0.83**	**0.77**
IMSI	0.41**	**0.70**	0.65	**0.72**	0.60*

Note: $r > 0.70$ are boldface. Correlations significant at the 0.01 level ($P < 0.01$) are without superscript; * = $P < 0.05$; ** = correlation is not significant.

Table 4.5 Regression analysis (R^2) between volume normalized OP at the sampling locations with the single species pollutants

	NR DRIVE (N = 18)	NDO (N = 20)	NDI (N = 21)	FDO (N = 17)	FDI (N = 17)
BC	0.19	0.31	0.19	0.08	0.11
CO	0.23	0.45	0.32	0.25	0.54
NO_x	0.18	0.44	0.52	0.20	0.34
$PM_{2.5}$ mass	0.37	0.32	0.55	0.70	0.60
EC	0.37	0.32	0.55	0.70	0.60
OC	0.66	0.72	0.68	0.63	0.44
IMSI	0.17	0.49	0.42	0.20	0.36

Table 4.6 Dorm indoor-to-outdoor Pearson Product Moment correlation (r) for each species. Note: $r > 0.70$ are boldface. Correlations significant at the 0.01 level ($P < 0.01$) are without superscript; * = $P < 0.05$; ** = correlation is not significant.

	Near Dorm	Far Dorm
OP	0.60 (N=22)	**0.88 (N=22)**
$PM_{2.5}$	0.62 (N=23)	0.60 (N=24)
OC	0.42* (N=24)	**0.76 (N=23)**
EC	0.17** (N=24)	**0.88 (N=23)**
CO	**0.98 (N=24)**	**0.76 (N=20)**
NOx	**0.99 (N=24)**	**0.99 (N=19)**
BC	0.17** (N=24)	**0.88 (N=23)**
IMSI	**0.97 (N=20)**	**0.98 (N=17)**

4.4. Discussion

The IMSI values exhibit similar seasonal trends compared to ambient CO, BC, and NOx with an increase in monthly average during the fall DRIVE study (Figure 4.2). Measurements from the NR GIT site and the UB site in 2015 show similar trends with the yearly minimum in the summer and increasing concentrations in the fall. At both near-road sites (NR DRIVE and NR GIT), the concentrations measured were low as compared to prior observations, highlighting the impact of decreasing vehicle emissions. These results were consistent with near-road measurements across the United States in 2015 (DeWinter et al., 2018). While vehicle emissions are still a major source for measured concentrations in the near-road environment, regional sources and meteorological conditions are increasingly important for driving concentrations in the near-road environment. Further, steep spatial gradients can lead to significant errors in estimated personal exposure using single pollutant measurements from near-road monitoring sites (D. H. Liang et al., 2018). In addition to the overall spatial variability observed, the concentration gradients as well as the correlations between the sites varied diurnally depending on the pollutant. Therefore, there are limitations to using single pollutant measurements in the near-road environment as a proxy for exposure to vehicle tailpipe emissions. Different exposure assessment strategies should be evaluated for understanding population exposure in the near-road environment.

The IMSI is based on three normalized pollutant concentrations and the mobile source contribution for the CO, NOx, and BC pollutant concentrations. While the spatial gradient and the diurnal profile for three pollutants were similar leading to an expected multipollutant profile (Figure 4.4), the multipollutant metric was able to better reflect the

spatial gradients of the tailpipe mobile source impacts by adjusting the normalized species concentrations by the contribution of mobile emissions to total emissions of that species. Further, when the multipollutant metric was assessed for significant driving factors in a multivariate regression, fewer terms were significant when compared to the single pollutants. This suggests that the IMSI is a more stable way to assess exposure compared to using a single pollutant measurement in the near-road environment.

Further, the metric is calculated based on a single site measurement; however, differences between the pollutant concentration levels at the NR DRIVE and the NR GIT sites highlight how site placement can affect concentrations. Therefore, concentrations from a single site could limit the accuracy of using either a single pollutant proxy or the IMSI metric for exposure of a population living in the near-road environment within a large urban area. Based on the assessments conducted here comparing sites within 2km of each other, the IMSI multipollutant metric can provide additional insight for characterizing exposure to primary traffic emissions. However, this evaluation is limited due to the temporal length of the study. Additional years of similar measurements may be able to provide additional understanding about how the IMSI metric can be used to better quantify exposure. Further, since the IMSI is an integrated metric using three pollutant concentrations, hourly data completeness of each pollutant can affect the number of hours the IMSI can be developed. As concentrations for pollutants continue to decrease in the near-road environment, it may be necessary to consider deriving a different equation for a statistical emissions-based metric.

The oxidative potential results were consistent with previous studies that showed that a significant source for water-soluble oxidative potential is secondary atmospheric

processing, including the processing of primary vehicle emissions. Volume normalized oxidative potential is related mainly to oxidized aromatic species, specifically PAHs oxidized to quinones and hydroxyquinones, and water-soluble transition metals, such as copper and magnesium (Verma et al., 2014; Verma et al., 2015). Both precursors are emitted from vehicles. Incomplete combustion is a source of PAHs and brake wear, tire wear, and resuspended road dust are sources for water-soluble transition metals. Oxidation of primary PAH emissions and acid dissolution of emitted metals is necessary to form water soluble species that become DTT-active (Cho et al., 2005; Fang, Guo, Verma, Peltier, & Weber, 2015; Meskhidze, Chameides, Nenes, & Chen, 2003; Nenes et al., 2011; Rattanavaraha et al., 2011). While the precursors are emitted as primary vehicle emissions in the near-road environment, oxidative potential of the water-soluble fraction of $PM_{2.5}$ measured with the acellular DTT assay does not appear to be a good mobile source-specific indicator. When considering both water-soluble and water-insoluble fractions of $PM_{2.5}$, the DTT assay captures a spatial trend in the oxidative potential of the fine particulate matter (Gao et al., 2017).

4.5. Conclusion

Characterizing exposure to primary mobile emissions using near-road monitoring becomes increasingly difficult as vehicle emissions decrease and are no longer the dominate contributing source to measurements. This study assessed two different multipollutant indicators as alternative surrogates of exposure to a single primary pollutant source. The Integrated Mobile Source Indicator does not overall improve the correlations between the sites, however it provides more consistent correlations spatially which could

help reduce exposure measurement error in future epidemiological studies. The IMSI can help here as it does utilize multiple species with differing pollutant levels above the background. The oxidative potential of fine particular matter exhibited a homogeneous spatial gradient linked to secondary components as opposed to primary emissions. While oxidative potential of water-soluble $PM_{2.5}$ constituents is potentially a good measure for overall exposure and fine particulate matter toxicity, it did not appear to be a good measure of exposure for primary vehicle tailpipe emissions.

CHAPTER 5

APPLICATION OF THE R-LINE DISPERSION MODEL RESULTS FOR CHARACTERIZING TRAFFIC-RELATED AIR POLLUTANT DYNAMICS FOR NEAR-ROAD EXPOSURE

Vehicular emissions are linked to several adverse health effects; however, quantifying exposure to the single source is increasingly difficult as vehicle tailpipe emissions continue to decrease. Dispersion models simulate spatially and temporally-resolved concentration fields from single source, mobile emissions. As part of a larger study aimed to understand exposure of individuals in the near-road environment, an intensive measurement campaign collected traffic-related air pollutants (TRAPs) and related data (e.g., meteorology, traffic, regional air pollutant levels) in Atlanta along one of the busiest highway corridors in the US. The near-road, fine-resolution model results were evaluated using the concentration measurements collected within the domain. Model concentrations were consistently biased low and high depending on the time of day. Different calibration methods were used to correct the model bias and improve the correlation between the measured concentrations. Accurate simulated concentrations in the near-road environment could improve exposure assessments to primary traffic-related air pollutants for populations in the near-road environment.

5.1. Introduction

Vehicle emissions are a major source of traffic-related air pollutants in urban areas despite emissions decreases due to improved controls and fuel regulations. Further about a fifth of the United States population lives within 500m of a heavily trafficked roadway and almost half of the population lives within urban centers (Rowangould, 2013). TRAPs have been linked to several adverse health effects emphasizing the need for source-specific exposure assessments (HEI, 2009c).

Personal monitoring, ambient measurements, and model concentrations are used in exposure assessments to traffic-related air pollutants; however, limitations with all models exist. Personal exposure monitoring accurately quantifies total exposure to ambient pollutant levels limiting the effectiveness of the method for determining exposure to a single source. Further, personal exposure monitoring can be both labor intensive and costly with limited individuals involved. Ambient air quality monitoring networks are often sparse in urban centers limiting spatially accurate exposure assessments. Further, urban monitoring locations are commonly placed in urban background locations to avoid direct influence from a single source and therefore do not capture the elevated concentrations found in the near-road environment. To help characterize TRAPs in the near-road environment, the US EPA implemented the Near-road Monitoring Network in 2014, which included up to two monitoring locations in 56 urban centers. The monitoring locations were placed along the busiest highways around the nation within 50m of the highway. These monitors provide near-road concentrations, but a single monitoring location cannot capture the small-scale spatial gradient in the near-road environment. While vehicle emissions are still a major single source in the near-road environment contributing to the concentration

of TRAPs, other urban sources and regional transport are becoming a more significant contributor to measurements in the near-road environment. The fraction of the pollutant concentration contributed by the mobile emissions is therefore unknown. Previous studies have included proximity to roadways or vehicle count as surrogates for TRAP emissions and as a result population exposure. As vehicle emissions decrease, the driving factors for temporal variably are no longer the surrogates assumed to represent the temporal variability of emissions. Further, the distance from the highway with the steepest portion of the concentration gradient decreases limiting the area in which the concentrations measured at the ambient near-road monitors are representative.

Using an air quality modeling system develops a complete spatially and temporally resolved concentration field for exposure assessments. Models use source emissions inputs and meteorological conditions to simulate source specific impacts at varying spatial scales. Chemical transport models are often used on the regional scale to simulate large-scale transport and transformation of primary and secondary pollutants. Computation requirements limit the minimum spatial resolutions to 4k. These models can therefore not capture the spatial gradients in the near-road environment. With simplified solvers and the lack of atmospheric chemistry, dispersion models can simulate primary pollutant concentrations at a finer resolution to capture gradients within an urban area. The Research Line (R-LINE) source dispersion model is a steady-state Gaussian plume model developed to simulate primary pollutant concentrations from line sources (Snyder et al., 2013; Venkatram, Snyder, Heist, et al., 2013).

The R-LINE model has been evaluated based on comparisons with other dispersion model simulations or based on experimental tracer gas concentrations (Heist et al., 2013;

Venkatram, Isakov, Pankratz, Heumann, & Yuan, 2004). Model characterizations show accuracy in the model predictions for simplified, short-term systems or dispersion from a single roadway (Venkatram, Snyder, & Isakov, 2013). These studies enable improvements to the model for certain meteorological conditions such as stable, low-wind conditions (Venkatram, Snyder, Heist, et al., 2013) or for taking into account site-specific characteristics include noise barriers. In order to apply dispersion models in epidemiological studies, the model performance evaluation needs to also be conducted in urban spatial scales considering the complexity of multiple roadway emissions (Batterman, Ganguly, & Harbin, 2015; Chang et al., 2015; Patton, Milando, Durant, & Kumar, 2017) and on different temporal scales in order to consider daily (Ganguly et al., 2015) as well as annual (X. Zhai et al., 2016) exposure. Previous studies have shown that R-LINE estimated annual averages relatively well with some slight overestimation. Discrepancies however have been attributed to limitations when attempting to estimate concentrations close to sources (Holmes & Morawska, 2006). When assessing the R-LINE daily and hourly average output, the model seems to fall short in capturing the variability that occurs over the course of seasons and days respectively.

Model evaluations conducted on the urban-scale include correlations with ambient measurements. In order to compare the single mobile source estimates with the observations, the background concentration or contribution to the ambient concentrations by all other sources needs to be included with the estimated concentrations or removed from the ambient measured concentrations. Continued assessments of these methods are needed for different urban areas and time scales to improve exposure assessments in order to continue characterize the health impacts of traffic-related air pollutants and drive

regulations on vehicle emissions. This study evaluates the R-LINE dispersion model to develop fine-scale near-road dispersion fields for hourly location-based exposure assessments. A regression approach was used to calibrate the R-LINE estimated concentration results to observations.

5.2. Methods

R-LINE dispersion fields were generated to evaluate the application of hourly dispersion modeling in traffic-related air pollutant exposure assessments for populations in the near-road environment. Pollutant concentrations measured at sites with varying distances from a major highway in Atlanta, GA were used to evaluate how well the R-LINE dispersion model captures the spatial and temporal variability of TRAPs in the near-road environment.

5.2.1 Mobile source dispersion modeling

The link-based 2010 emissions inventory was provide by the Atlanta Regional Commission's (ARC) 20-county activity-based travel demand model and mobile source emissions modeling. The road network consisted of 43,712 links over the 20-county regional area surrounding Atlanta, GA and was updated to 2014 by scaling with the ratio of the annual 2014 to annual 2010 average emissions levels (NOx: 0.69 and CO: 0.81) from the Motor Vehicle Emissions Simulator (MOVES) (EPA, 2010b). ARC generated hourly link-based emission rates for an average weekday diurnal profile (Figure 5.1) through the Atlanta Roadside Emissions Exposure Study (AREES) in order to quantify traffic impacts on local air quality in Atlanta (D'Onofrio, 2015). Since diurnal meteorological conditions

and traffic patterns are correlated, using daily average emissions would have led to elevated simulated concentrations by R-LINE and therefore the average weekend diurnal tailpipe emissions profile was used (X. Zhai et al., 2016). The link-based emission factors (gram vehicle^{-1} mile^{-1}) are based on link specific information including road type and location, AADT, and fleet demographics.

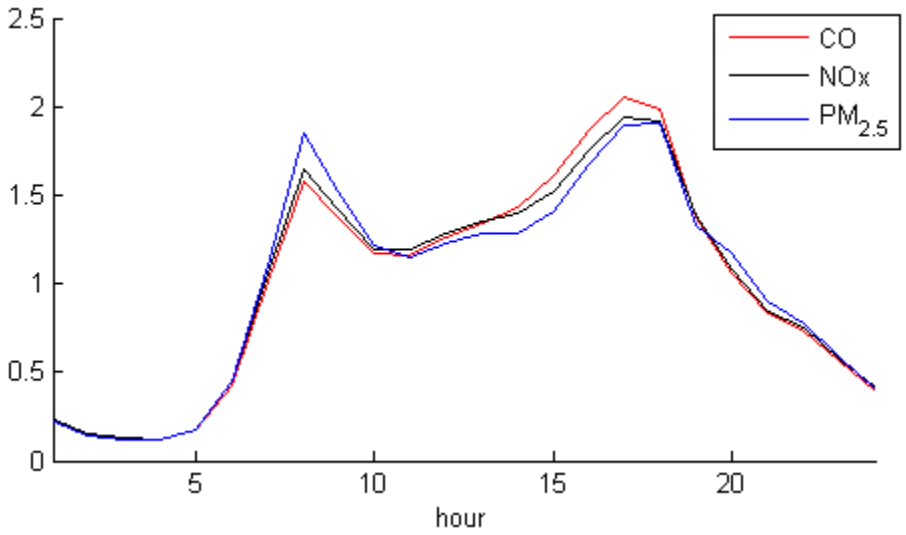

Figure 5.1 Diurnal profile of tailpipe emissions normalized by mean weekday emissions generated during AREES by ARC

Surface meteorological data was from the National Weather Service (NWS) site at the Hartsfield-Jackson International Airport (KATL) and the upper air data was from the Peachtree City Falcon Field Airport (KFFC). The hourly meteorological data input for R-

LINE was processed using AERMET (version 15181) to produce the additional variables necessary for R-LINE including the surface friction velocity, convective scale, Monin-Obukhov length, and surface roughness (Cimorelli et al., 2005). Wind speed below $1ms^{-1}$, which represented three percent of the hours, were changed to $1ms^{-1}$ according to EPA recommendations to reduce over prediction common during low wind conditions (EPA, 2000).

Primary concentrations of CO and NOx from on-road mobile sources were simulated using the steady-state dispersion model Research LINE (R-LINE) source (version 1.2) (Snyder et al., 2013). R-LINE was run using the default numerical integration method and the other default conditions were used with an error limit of 0.001. The beta option for roadside barriers and depressed roadways was not used.

5.2.2 Exposure Domain

This study focused on traffic-related air pollutants emitted from the segment of highway where Interstate 75 and Interstate 85 (I-75/I-85) have merged in the center of Atlanta, Georgia (Figure 5.2). The highway supports an annual average daily traffic (AADT) of about 330,000 vehicles of primarily light-duty gasoline passenger cars and trucks. Heavy-duty diesel trucks make up approximately four percent of the total AADT on this segment of the highway. A 1.1km segment of the highway passes through or boarders the east side of the Georgia Institute of Technology (GIT) campus. The dispersion modeling study domain was 6.5km by 2.5km centered on the GIT campus with a receptor grid resolution of 25m (24,341 receptors). The simulated concentration domain was also

centered within the emissions factors input domain to ensure the inclusion of link-based emissions occurring outside the R-LINE modeled domain.

Figure 5.2 Model Domain included monitoring locations. NR GIT: Near-road Monitoring Network monitor on the GIT campus; NR-DRIVE: Near-road DRIVE site; ND: Near highway dorm outdoor and indoor sampling; FD: Far dorm outdoor and indoor sampling; UB: Urban background Jefferson St SEARCH site

5.2.3 Monitoring Data and Model Calibration

The study domain over the Georgia Institute of Technology campus contained five monitoring sites used for model evaluation. The Dorm Room Inhalation to Vehicle Emissions (DRIVE) study monitored traffic-related air pollutants at three sites of varying distance from I-75/I-85 for September 8, 2014 to January 5, 2015. The near-road sampling

site (NR DRIVE) was located 6m from the west side of the highway in an open parking lot with less than 85 passenger vehicles. The CO (Thermo Model 48i), NOx (Teledyne API 200A), and BC (Magee Aethalometer AE31) sampling inlet height was 3m and about 6m west of the closest highway lane. The two sites in student dormitories were located 20m and 1.4km from the highway and measured CO (Teledyne 300E), NOx (Thermo 42C), and BC (microAeth AE51). The site closer to the highway (Near Dorm or ND) operated out of an occupied administrative office on the ground floor of a five-story building and had an inlet height of 0.5m. The site further from the highway (Far Dorm or FD) operated out of an empty room part of a two bedroom-one bathroom suite on the ground floor of a five-story building and had an inlet height of 1.5m.

An additional near-road site (NR GIT) was part of the EPA Near-road Monitoring Network located on the GIT campus. This site is located within a row of trees and a limited-access parking lot borders the site. The urban background (UB) site is part of the Southern Aerosol Research and Characterization (SEARCH) network located 2.3km west of the highway. Previous studies have assessed this site as representative of Atlanta urban background pollutant concentration and composition (Edgerton et al., 2005; Liu et al., 2005; Solomon et al., 2003). The concentration data for the NR GIT site and the UB site were obtained from the US EPA Air Quality System and the SEARCH network (Hansen et al., 2003).

Hourly CO and NOx concentrations estimated by R-LINE were calibrated with monitoring data at five measurement locations. Previous research has shown that locally elevated simulated concentration levels co-occur with specific meteorological scenarios (Perry et al., 2005; Venkatram, Snyder, Isakov, & Kimbrough, 2013; Venkatram, Snyder,

Heist, et al., 2013). In addition, unrealistically high concentrations were observed on and very near the main highway emissions source. In order to correct for overestimation, a number of processes to adjust the data are compared. Three calibration methods were applied by developing linear, diurnal linear, and nonlinear regressions based on hourly comparison between observed and modeled concentrations.

5.2.4 Model evaluation metrics

The fields developed for this study were to support the objective of the DRIVE study to assess spatial and temporal variability for epidemiological studies focusing on hourly exposure to populations in the near-road environment. Specifically, as individuals on the GIT campus move throughout the day between their dormitories and academic buildings, their exposure to traffic-related air pollutants will vary based on time of day and distance from the highway (Liang, 2018). Previous R-LINE evaluations consider either daily or annual averages since these averaging periods are commonly used in residential-based health impact studies (Heist et al., 2013; Snyder et al., 2013; Venkatram, Snyder, & Isakov, 2013; X. Zhai et al., 2016). Here, hourly-simulated concentration fields were evaluated since the desired application was to estimate location-based traffic-related personal exposures. Analyses were conducted based on pollutant, wind direction, monitoring site, and time of day. Statistical evaluations of the modeled results to the measured concentrations included the root mean square error (RMSE) metric and the Spearman correlation coefficient.

5.3. Results

Average concentration fields of the hourly average CO and NOx concentrations were developed by R-LINE for the study domain focusing on the Georgia Institute of Technology campus. With an AADT 15 times greater than the surrounding surface streets, the interstate is the major source within the domain. Estimated concentrations decrease with distance from the I-75/I-85 interstate as expected and observed in the measurements (Table 5.1).

5.3.1 Model estimates

For CO, the average hourly concentrations at the NR DRIVE and NR GIT ambient measurement sites were 425 ppb and 624 ppb, respectively (Table 5.1). In comparison, the average hourly CO modeled concentration at the NR DRIVE and NR GIT sites were 1073 ppb and 945 ppb. For NOx, the average hourly concentrations were 50 ppb and 57 ppb, respectively at the NR DRIVE and NR GIT sites (Table 5.1). The average hourly NOx modeled concentration at the NR DRIVE and NR GIT sites were 134 ppb and 117 ppb. While the modeled average near-road concentrations are within a factor of about two, the modeled concentrations represent the concentration contribution of only a single source and were expected to be lower than the observations. While the model largely overestimated concentrations within 20m of the highway, estimated concentrations are also bias high with increasing distance from the highway. At the urban background site 2km from the highway, the average model estimates are a factor of about 1.5 times greater than the observations.

Table 5.1 Site mean observations and R-LINE estimates for September 1 to December 31, 2014

Pollutant	Site	Hours	Obs	N	Model	Rsp	RMSE	Background
NOx	NR DRIVE	2666	50	2906	134	0.55	31	-66
	NR EPD	2798	57	2906	117	0.35	32.5	-47
	NDO	2357	39	2906	95	0.57	32	-34
	FDO	1883	33	2906	57	0.43	29.2	-50
	UB	2627	28	2906	43	0.61	31.9	-1
CO	NR DRIVE	2178	425	2906	1078	0.62	187	-494
	NR EPD	2816	624	2906	945	0.42	288	-192
	NDO	2226	344	2906	776	0.47	160	-203
	FDO	1914	204	2906	468	0.41	178	-473
	UB	2648	268	2906	354	0.58	180	57

Abbreviations: Obs = Overserved concentrations; Model = Modeled traffic impact; Rsp = Spearman's correlation coefficient; NRMSE = Normalized root mean square error; Background = Calculated background concentration

Maximum hourly average concentrations near the highway were largely overestimated with CO and NOx simulated concentrations within the domain of 73556 ppb and 8717 ppb, respectively. Within about 50m, the concentration drops drastically suggesting the near-road gradients are too steep. Maximum hourly average concentrations at the modeled receptor grids located at the NR DRIVE and NR GIT site locations for CO were 30280 ppb and 31510 ppb, respectively and for NOx were 3861 ppb and 4024 ppb, respectively. The maximum hourly concentrations for CO measured at the NR DRIVE and NR GIT sites were 1860 ppb and 2200 ppb, respectively, and maximum NOx concentrations were 201 ppb and 233 ppb, respectively. While site location differences between the NR DRIVE and NR GIT sites lead to different near-road measurements, the R-LINE estimates were higher than the near-road observations by a factor of about 15 for CO and about 18 for NOx. For comparison, the maximum concentrations simulated and

measured for CO at the urban background site were 20641 ppb and 1389 ppb, which is a factor of 15. Similarly, the maximum concentrations simulated and measured for NOx at the urban background site were 2650 ppb and 280 ppb, which is a factor of about nine. The overestimated CO and NOx concentration across the whole domain suggests the emissions input file or dispersion from the roadway may be driving this high bias.

The diurnal profile for the observations shows a peak in the concentration for CO and NOx in the morning around 9am and in the evening around 7pm. While highways dominated by work commuters observe a daily bimodal trend in traffic count, the traffic count on the main highway in the domain remains high from 6am to 8pm. The drop in concentration observed in the average diurnal profile is likely driven more by daily meteorology as the mixing height increased during the afternoon. The CO and NOx concentrations simulated by the R-LINE model follow a similar bimodal concentration trend, however the estimated concentrations are a factor of 3.2 during the morning and evening peaks and a factor of 0.4 the average ambient concentrations during the afternoon minimum. This diurnal profile trend remains consistent for CO and NOx with increasing distance from the highway; however, the concentration spread between the morning maximum and afternoon minimum increases with distance. At the UB site, the morning peak is a factor of 4.1 above the average and the afternoon minimum is a factor of 0.2 below the average.

This evaluation of R-LINE assessed the dispersion model performance in relation to hourly average concentrations of CO and NOx at multiple sites within the near-road environment in Atlanta, GA. While the results did not identify R-LINE as capable of capturing temporal variability observed at the five monitoring locations, different

calibration methods showed improvements in the model evaluation. Although R-LINE estimations were bias high, the spatial gradient observed with the 25m receptor grid resolution showed the traffic pollutants reaching urban background levels within 50 – 100m from the edge of the nearest highway lane. This spatial gradient is consistent with previously reported distances (Beckerman et al., 2008; Boogaard et al., 2011; Kozawa, Winer, & Fruin, 2012). Further, observations within the domain showed the steepest gradient to be within 60m of the nearest highway lane between the DRIVE near-road site and the near dormitory site. Differences between the NR DRIVE and NR GIT sites suggest that sampling location does contribute to the near-road concentration; however, the R-LINE dispersion modeling results are qualitatively consistent with the ambient monitoring showing the sharp pollutant concentration reduction over the relatively short distance from the highway. Observations showed and the R-LINE dispersion modeling confirmed further reductions in CO and NOx concentrations continue with increased distance.

Capturing personal exposure based on mobility throughout the day requires accurate hourly concentration data. The use of ambient measurements from a single sampling location leads to measurement error due to spatial variability around the monitor as well as temporal variability in the correlation between two locations (D. H. Liang et al., 2018). While additional monitoring locations could help improve the spatial resolution of pollutant concentrations, spatially-resolved concentration fields from modeling can improve characterization of temporal exposure trends. Accurate hourly concentration fields provide complete resolution in order to follow individual location for more sensitive indicators of personal exposure. A combination of calibrated R-LINE dispersion modeling

and GIS location tracking information could improve personal exposure prediction performance to traffic-related air pollutants.

5.3.2 Model calibration and evaluation

This analysis evaluated the relationships between the R-LINE estimates and the observed concentrations at five monitoring locations within 2km of a major highway using linear and non-linear regressions. To consider the robustness of the comparison, the regression methods were also evaluated using only the NR GIT and UB sites. The comparison indicates that the R-LINE estimates poorly capture the CO trends at the monitor locations with linear comparison Pearson (R^2) correlation coefficient values and Spearman (r_s) correlation coefficient values of 0.11 and 0.52, respectively. The comparison for NOx also indicates that the R-LINE estimates poorly capture the trends at the monitor locations with linear comparison Pearson (R^2) correlation coefficient values and Spearman (r_s) correlation coefficient values of 0.16 and 0.57, respectively.

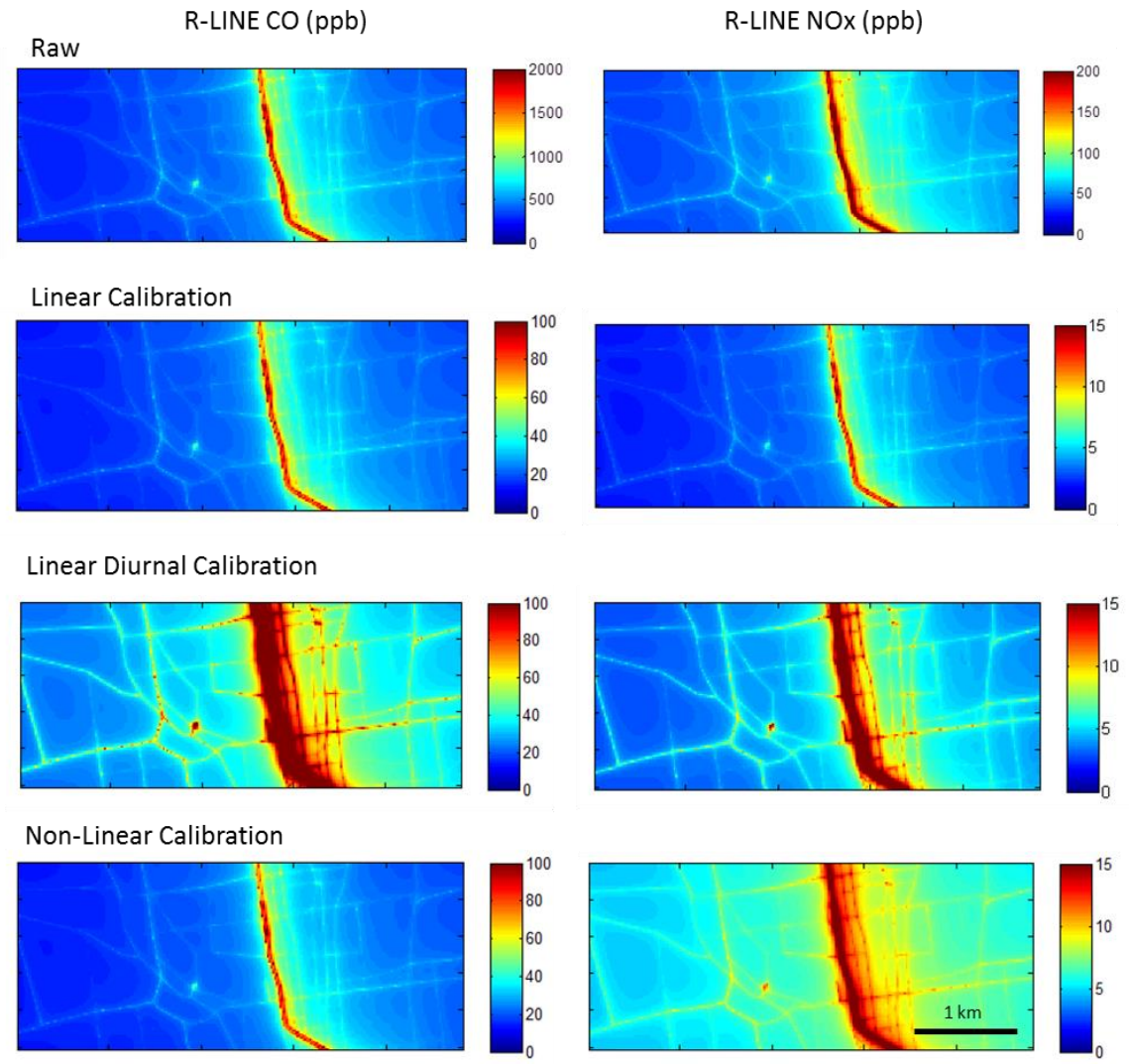

Figure 5.3 Hourly averages of R-LINE estimates by mobile sources from September 8, 2014 to December 5, 2015.

5.3.2.1 Linear Calibration

Due to the overestimation of the R-LINE model, simulated fields of the CO and NOx species are biased high compared to the observational data. The slopes of the linear regression for the R-LINE estimated and the observational data were 0.05 and 0.06 for CO

and NOx respectively. The slopes of the linear regression for the R-LINE estimated and observational data when only using the NR GIT and UB sites were 0.06 and 0.06 for CO and NOx respectively. While additional monitoring location data were available during the study period, the linear calibration evaluation would be similar outside the DRIVE study period using fewer observational data sites. The linear regression intercept was evaluated as the background concentration or the concentration with no mobile source contribution. The intercept for the linear regressions for CO and NOx were 343 ppb and 33.7 ppb, respectively. These background concentrations are higher than observations at a rural site northeast of Atlanta, however still reasonable (Blanchard, Hidy, et al., 2013a). While using a linear calibration does not impact the Pearson (R^2) correlation coefficient values and Spearman (r_s) correlation coefficient values, the maximum hourly average concentrations within the domain were largely reduced for CO and NOx to 3517 ppb and 503 ppb, respectively.

Table 5.2 Site observations and linear calibration R-LINE estimates for September 1 to December 31, 2014

Pollutant	Site	Hours	Obs	N	Model	Rsp	RMSE	Background
NOx	NR DRIVE	2666	50	2906	7.7	0.55	31	54
	NR EPD	2798	57	2906	6.8	0.35	32.5	60
	NDO	2357	39	2906	5.5	0.57	32	37
	FDO	1883	33	2906	3.3	0.43	29.2	44
	UB	2627	28	2906	2.5	0.61	31.9	22
CO	NR DRIVE	2178	425	2906	51.5	0.62	187	399
	NR EPD	2816	624	2906	45.2	0.42	288	677
	NDO	2226	344	2906	37.1	0.47	160	332
	FDO	1914	204	2906	22.4	0.41	178	289
	UB	2648	268	2906	16.9	0.58	180	230

A few factors may drive the over estimation by the R-LINE model when compared to the near-road monitoring locations including the formulation of the model or uncertainties in the emissions input. The R-LINE model does not include chemical reactions or wet deposition as a species sink. While CO is a relatively stable primary pollutant with low reactively and deposition loss, NOx is readily lost through atmospheric reactions. Since this evaluation is based on the hourly accuracy of the R-LINE model, it was important to compare the average diurnal profiles. The CO and NOx concentrations simulated by the R-LINE model follow a similar bimodal concentration trend, however the estimated concentrations are a factor of two less than the average ambient concentrations once the background (i.e. linear calibration intercept) is added to the regression-adjusted estimated concentrations.

RLINE develops concentration estimates based on only mobile source emissions while observations are total ambient concentrations, which may lead to some of the diurnal

variability. Hourly concentration fields however could substantially improve personal exposure assessments. Calibrating the R-LINE model concentration estimates with observations is a possible method to improve model estimates to more accurately characterize the mobile source impacts for traffic-related air pollutant exposure and regulations. As vehicle emissions continue to drop with improved engine technology and fuel regulations, it becomes more difficult to use near-road monitoring to quantify the source contribution of mobile source to ambient species concentrations.

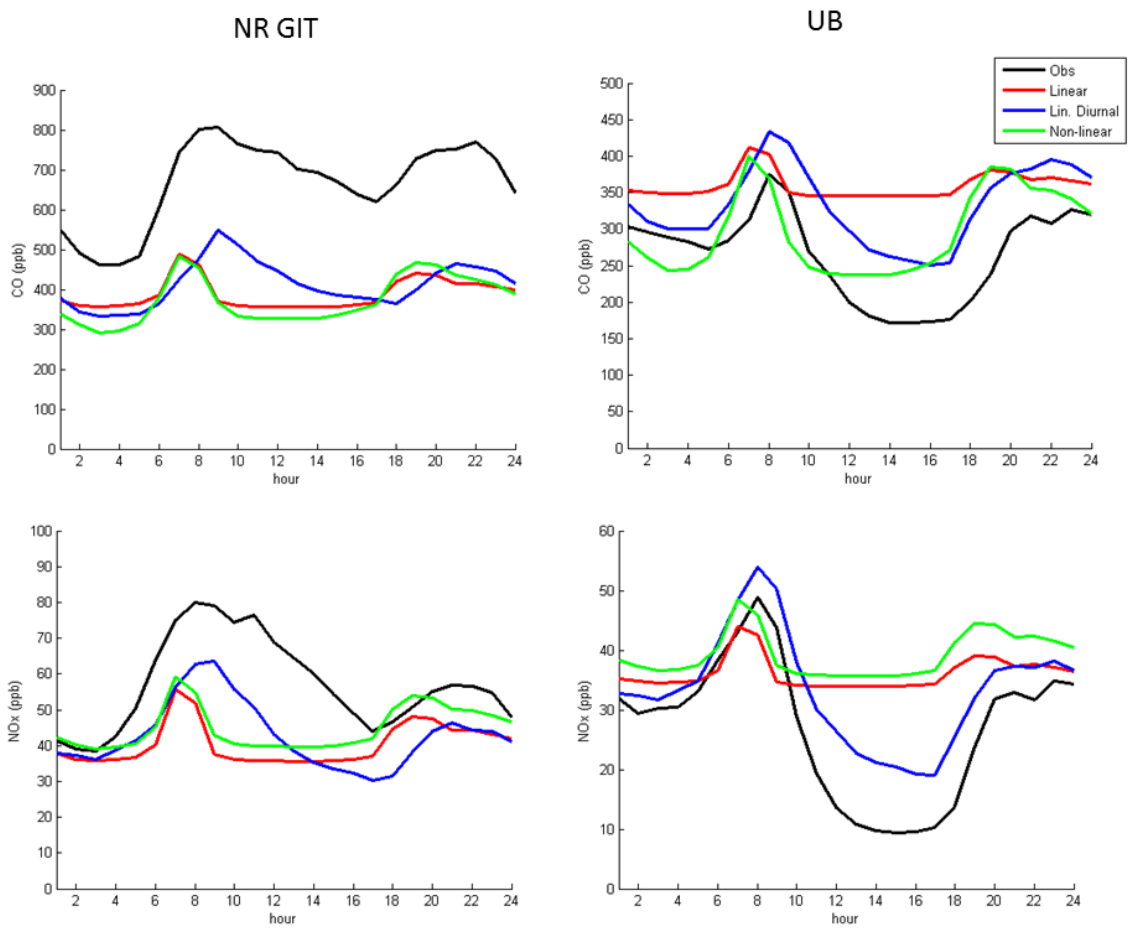

Figure 5.4 Diurnal profile for a near-road site and the urban background site. Calibration methods for R-LINE estimates include background added to hourly concentrations.

5.3.2.2 Diurnal Linear Calibration

While the linear calibration reduced the biased high concentrations, it did not impact the shape of the diurnal profile. By applying the diurnal linear calibrations, the slope varied between 0.03 and 0.6 for CO with the intercept range of 224 ppb to 392 ppb. The slopes for NOx had similar ranges from 0.04 to 0.7 with an intercept range of 16 ppb to 46 ppb. By allowing the slope and intercept to vary, the shape of the diurnal profile could

adjust to follow a similar tread as the observed diurnal profile (Figure 5.4). While the profile at the UB site closely matched the observations with a slight increased offset ranging from 10 ppb at 2am to 100 ppb at 2pm for CO and 2 ppb at 1am to 12 ppb at 1pm for NOx, the calibrated model estimated at the NR GIT site showed a greater offset for CO and NOx below the observations. For the normalized profiles the UB site followed the observation trends with the morning peak a factor of about 1.5 the average and the afternoon minimum at the average, the diurnal linear calibrations applied to the near-road sites were not able to adjust the trends to follow the observation profile (Figure 5.5). The diurnal linear calibrations improves the ability for the R-LINE model to capture the observations at the monitor locations with a Pearson (R^2) correlation coefficient value and Spearman (r_s) correlation coefficient value of 0.21 and 0.48 for CO and 0.23 and 0.51 for NOx, respectively.

Table 5.3 Site observations and linear diurnal calibration R-LINE estimates for September 1 to December 31, 2014

Pollutant	Site	Hours	Obs	N	Model	Rsp	RMSE	Background
NOx	NR DRIVE	2666	50	2906	17.1	0.45	30.4	38.6
	NR EPD	2798	57	2906	13.5	0.4	30.8	48.8
	NDO	2357	39	2906	9.5	0.46	30.5	30.5
	FDO	1883	33	2906	5.4	0.31	28.5	42
	UB	2627	28	2906	3.9	0.46	30.3	-1
CO	NR DRIVE	2178	425	2906	144	0.47	187	252
	NR EPD	2816	624	2906	112	0.41	276	561
	NDO	2226	344	2906	79	0.48	149	266
	FDO	1914	204	2906	44	0.39	170	243
	UB	2648	268	2906	31	0.45	168	57

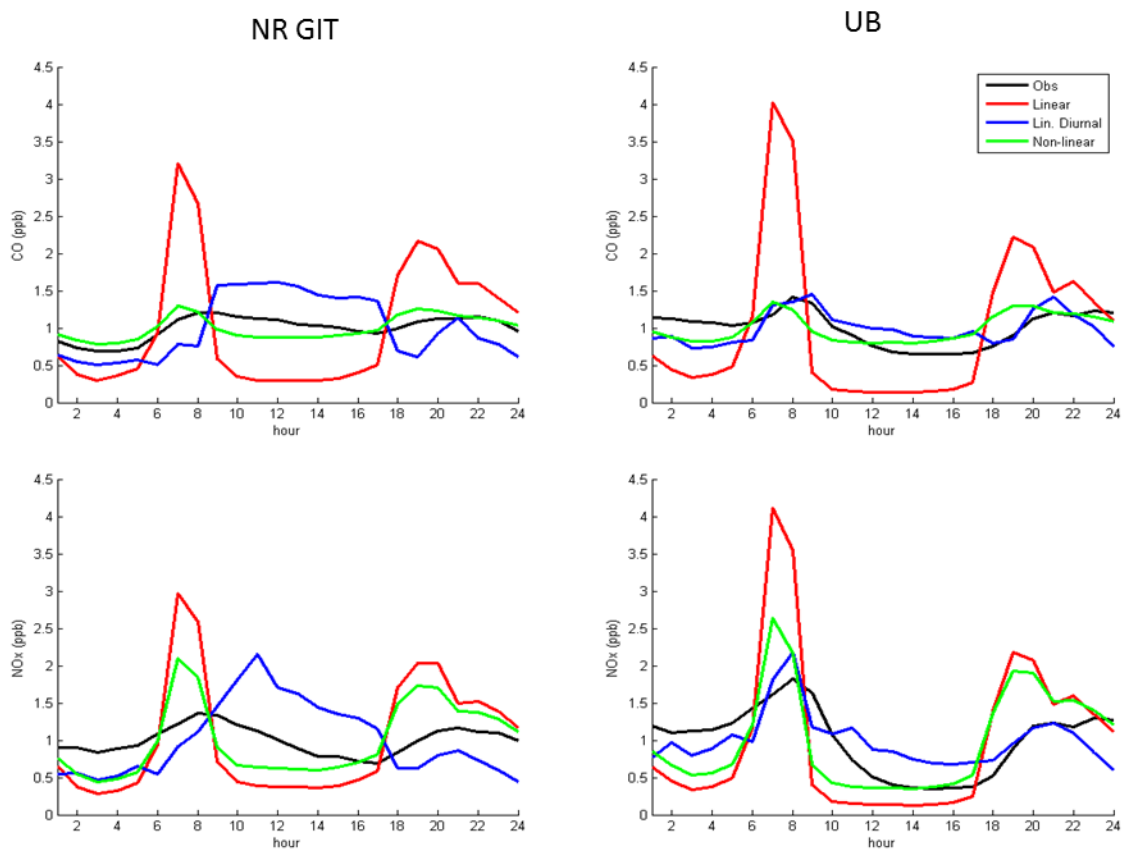

Figure 5.5 Normalized diurnal profile by mean for a near-road site and the urban background site. Calibration methods for R-LINE estimates do not include background added to hourly concentrations.

In addition to high concentrations generated closer to roadways, R-LINE results had strong concentration peaks at 7AM and 6PM and concentrations close to zero between 10AM and 3PM for all species. While the diurnal profiles for the measured pollutant concentrations also showed two clear peaks in the morning and evening, the drop in the middle of the day was not as dramatic. The third method will develop hourly linear regressions and explore why model produces this particular diurnal profile shape. By improving the accuracy of the R-LINE spatial and temporal results, the concentration fields

can more accurately represent personal exposure to primary roadway tailpipe emissions based on location tracking data.

5.3.2.3 Non-linear Calibration

The non-linear calibration has a similar overall correlation as the diurnal linear calibration. The non-linear comparison has a Pearson (R^2) correlation coefficient values and Spearman (r_s) correlation coefficient values of 0.23 and 0.52 for CO, and 0.19 and 0.57 for NOx, respectively. The CO and NOx concentrations simulated by the R-LINE model follow a similar bimodal concentration trend as the linear calibration diurnal profile, with the exception of the CO profile at the UB site which has a significant decrease in concentration from 9am to 6pm (Figure 5.4). The normalized profile for CO shows a lower maximum and higher minimum than the normalized linear calibration profile. For NOx, the normalized non-linear profile appears to follow the normalized linear calibration trend (Figure 5.5).

Table 5.4 Site observations and non-linear calibration R-LINE estimates for September 1 to December 31, 2014

Pollutant	Site	Hours	Obs	N	Model	Rsp	RMSE	Background
NOx	NR DRIVE	2666	50	2906	12.7	0.56	29.9	47.6
	NR EPD	2798	57	2906	11.3	0.35	32.2	54
	NDO	2357	39	2906	9.7	0.57	31.6	32
	FDO	1883	33	2906	7	0.43	29	38
	UB	2627	28	2906	5.6	0.61	30.2	19
CO	NR DRIVE	2178	425	2906	393	0.62	168	25
	NR EPD	2816	624	2906	373	0.42	274	315
	NDO	2226	344	2906	356	0.47	153	-10
	FDO	1914	204	2906	323	0.41	172	-87
	UB	2648	268	2906	296	0.58	164	-32

5.4. Conclusion

The R-LINE dispersion model hourly estimated traffic-related air pollutant concentrations are bias high in the near-road environment. Model formulations driving the diurnal profile trends do not follow the profile shown in the ambient observations. Basic linear and non-linear calibrations with measured concentrations are evaluated for improved spatial and temporal variability compared to ambient concentrations. Based on the overall Pearson and Spearman correlation coefficients, none of the calibration method improve the R-LINE model estimates. It is important however to also consider how the diurnal profile changes with the calibration method as well as the impact on the absolute concentrations. The linear diurnal calibration method leads to spatial gradients and temporal profiles that follows those observed in the ambient measurement data.

CHAPTER 6

CONCLUSIONS AND FUTURE WORK

6.1 Conclusions

The objective of the research conducted for this dissertation was to improve understanding of the near-road microenvironment for exposure assessments to tailpipe emissions and to assess methods for exposure analyses. Historically, emissions from vehicle tailpipes have contributed to elevated concentrations within a couple hundred meters of highly trafficked roads leading to the single dominate source contributing to pollutant measurements. Emissions from the single source could then rely on measurements in the near-road environment. The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study was designed to characterize the near-road environment across the Georgia Institute of Technology in Atlanta, GA from six monitoring sites and assess a range of exposure methods for vehicle tailpipe emissions. The study outline was described in chapter two in order to provide the background and justification for the measurements collected.

Physical site characteristics, such as natural and built barriers, can impact the concentrations measured leading to differences in measurements along the same highway segment. The bias observed between two sites located 300m from each other were described in chapter three to understand how the impact of vegetation around monitoring sites can impact the assessment of population exposure that relies on single species

measurements. Reduced dispersion at the site located within a tree line led to higher measured CO and NO concentrations as lower NO_2 concentrations on average compared to the site located in an open, asphalt parking lot.

Decreased vehicle tailpipe emissions from improved engine technology and fuel regulations have led to a lower contribution to concentrations measured in the near-road environment compared to other sources also emitting key traffic-related air pollutants such as CO, NOx, and $PM_{2.5}$ in the urban area. The traffic-related air pollutant concentrations measured at the near-road sites showed a reduced impact from the vehicle emissions on the highway with less elevated concentrations above the urban background compared to prior studies. These measurement observations indicate the effectiveness of mobile source emission controls leading to the decrease in contribution from vehicles to urban air pollution and a changing near-road environment that will impact future approaches used to characterize exposure to vehicle emissions.

A number of different methods are used to assess exposure to primary tailpipe mobile source emissions. In addition to single species tracers, several multipollutant metrics have been developed as alternative surrogates of exposure to primary mobile pollutants. Chapter four evaluated the usefulness of an emissions-based statistical metric and a biological-based metric. The emissions-based metric used was the Integrated Mobile Source Indicator (IMSI) and while it did not improve the correlation between the sites, it provided more consistent correlations spatially. Used as a metric for exposure in epidemiological studies, the IMSI could reduce exposure measurement error compared to single pollutant tracers since it applies three species with different concentration ratios above background. The biological-based metric used an acellular assay to measure the

oxidative potential of water soluble fine particulate matter components. The metric displayed a spatial gradient similar to OC and $PM_{2.5}$ mass suggesting it is linked to secondary $PM_{2.5}$ components more than primary mobile emissions components such as black carbon.

Dispersion modeling provides a simplified method to estimate the impact of single source emissions at any defined receptor grid. For exposure assessments to incorporate daily movement within the near-road environment, dispersion modeling provides hourly, spatially-resolved concentrations fields. Bias observed in the simulated concentrations for roadway vehicle tailpipe emissions is shown in Chapter 5 focusing on the differences between the simulated and observed diurnal profiles. Methods to reduce the bias included linear and non-linear calibrations, though these simple calibration processes did not improve the correlations between the observed and simulated concentrations.

6.2 Future Work

6.2.1 Near-road Measurements

The research conducted for this dissertation characterized the spatiotemporal dynamics of the near-road microenvironment around one of the busiest highways in the country showing an overall decrease in impact from vehicle emissions. While traffic count has been a dominant driving factor and surrogate for vehicle emissions impacts in the past, fleet turnover continues to reduce per-vehicle emissions, and other site specific factors drive pollutant dynamics observed at a monitoring site location. The EPA Near-road Monitoring Network provides an on-going set of monitoring sites to characterize temporal

trends in near-road pollutant concentrations as urban fleets continue to improve. Further assessment can also provide an insight as to how site location differences across the country impact measurements and what factors drive concentration trends. These concentration measurements reinforce the effectiveness of engine and fuel control policies in reducing overall emissions from mobile sources and can provide further insight into which pollutants should continue to be a focus. While nitrogen dioxide was the focus of the EPA Near-road Monitoring Network under the hypothesis that the concentrations would be greatest in the near-road environment, the concentrations were found to be below national standards in this microenvironment. Monitoring sites that were planned for implementation in the third phase of the program have already been cancelled as a result of the lower than expected concentrations. Future work with the active monitoring sites part of the near-road network should continue to assess concentrations in the near-road environment to further characterize the impact of monitoring siting. By focusing on temporal dynamics observed at specific sites, future work can be driven to understand the factors that can be used as surrogates where traffic count is not sufficient. This network of sites also creates a unique opportunity to better understand photochemical reactions and ozone titration in the near-road environment.

While the Near-road Monitoring Network provides temporal trends of the source impact to primary pollutants, the single monitoring location cannot provide information about the changing trends in the spatial gradients observed within a couple kilometers of major national highways. A single monitoring site requires a large amount of monetary and personnel resources. Each site requires a weather-proof shelter held at a consistent temperature and a range of instrumentation necessary for different air pollutants. Further,

maintaining the instrumentation for high data quality requires knowledgeable personnel and therefore appropriate training should be used to connect state employed personnel with local universities. Recognizing the resources required for monitoring sites is high, semi-permanent mobile sites could be an efficient use of capital in order to increase the dataset of near-road spatial gradient measurements. A handful of insulated trailers or vehicles could be placed in a temporary location for six months to a year to observe pollutant concentrations for a couple different seasons. Key pollutants would include nitrogen oxides, carbon monoxide, black carbon, ozone, particle composition, and ultrafine particles. Other key measurements would include traffic count and vehicle type, temperature, relative humidity, wind speed, and wind direction. Mobile measurements from vehicles on roadways have been used to identify hotspots from roadside sources or common delivery routes (Apte et al., 2017). While this method captures spatial gradients on roadways, these measurements are not able to provide insight on spatial pollutant concentration gradients perpendicular to roadways. Exposure to primary traffic-related air pollutants is highly dependent on local source contributions, meteorology, traffic patterns, and topography that impacts dispersion. Due to the local nature of spatial gradients, a network of temporary monitoring sites would develop several short-term databases around urban areas to characterize exposure to a greater percentage of the individuals working, commuting, and living within this unique microenvironment.

An improved understanding of the spatial trends in an urban core could help direct city specific plans to deal with TRAPs. Utilizing temporary monitoring sites to create a database of short-term measurements around an urban area could help identify the locations of high pollutant concentrations. The type of industries and the design of the street network

will impact the location of pollutant hotspots around different cities and these locations are not always intuitive. Utilizing non-federal reference method (FRM) instrumentation, such as low-cost sensors, could also contribute to the development of network used to highlight hotspot locations and assess spatial resolution. Low-cost sensor technology has improved considerably and will continue to improve for applications in future measurement networks. A mobile monitoring platform could help highlight these locations before limited resources are spent implementing a permanent monitoring site (Apte et al., 2017; Hankey & Marshall, 2015). Further, once these locations are identified and actions take place to reduce the emissions in the hotspot, the site may be better utilized in a different location to continue reducing key sources of urban emissions.

6.2.2 Multi-pollutant Indicators

Several of the findings throughout this dissertation, including the variability observed between the two near-road monitoring sites along the same road segment, highlighted the complexity that exists in the near-road environment. While the multipollutant indicators assessed had shown indications of being useful in characterizing exposure in the near-road environment, both indicated there were limitations and reinforced the complexity of this micro-environment observed in the primary pollutants that were measured.

Previous applications for the integrated mobile source indicator were on the urban scale (M. M. Oakes et al., 2014; Pachon et al., 2012). Here we applied the metric within the near-road environment unable to assess the metric with health data. Further comparison and assessment of different metrics at different spatial scales are needed in future work.

The IMSI has been assessed in three different urban locations, however the statistical multipollutant methods developed need to be assessed at different spatial and temporal scales as well.

The measure of oxidative potential for water-soluble $PM_{2.5}$ via the DTT assay showed a slight correlation with the single traffic-related air pollutants traditionally measured as markers of mobile emissions (BC, NOx, and CO). The correlations varied widely based on the pollutant and the rate of decrease in the correlation with distance from the highway was less than that observed in the pollutant measurements. Further assessment is needed to understand what factors drive or impact the oxidative potential of fine particulate matter. Atmospheric processing appeared to be an important factor required to process primary emissions into secondary species that drove the oxidative potential of the aerosols. The DTT assay is only one measure of oxidative potential and the literature utilizing this assay is limited. Oxidative potential is linked to adverse health effects and further evaluation using the DTT assay as well as other oxidative potential assays should continue. The oxidative potential data utilized for study was integrated, 48-hour filter samples. The use of integrated measurements limits the oxidative potential interpretation and the temporal correlation analysis. In addition, filter measurements can include positive or negative artifacts due to the length of the collection time. Automated, continuous measurement techniques would improve this limitation. Pollutant concentrations were measured continuously throughout the DRIVE study and averaged to match the oxidative potential filter measurements. Time resolved hourly correlations could improve the understanding on the factors that drive oxidative potential diurnally.

While prior research suggested that mobile sources are linked to reactive oxygen species in fine particulate matter, the oxidative potential of particles increases is found to increase with atmospheric processing. Therefore, the DTT assay can be used as an indicator of mobile source emissions when measuring total $PM_{2.5}$, but the species driving the potential health effects of traffic-related air pollutants may be secondary species from atmospheric processing of the traffic-related air pollutants. The oxidation of PAHs to quinones and hydroxyquinones as well as transition metals support that atmospheric processing is potentially important to the resulting health effects from fine particulate matter. The results from the DTT assay suggest that further work should be conducted to understand the atmospheric transformation processes that increase the oxidative potential activity of pollutant emissions and the impact of primary mobile emissions on secondary pollutant concentrations. Future work might address this by characterizing the chemical composition of particulate measurements in the near-road environment at the same locations where filters are collected for oxidative potential measurements to observe if a spatial gradient is observed in the particulate composition as well. This is increasingly important for the changing near-road environment were the influence of primary mobile emissions is becoming less pronounced.

6.2.3 Dispersion modeling

The spatially and temporally resolved concentration fields simulated with dispersion modeling provided an alternative method to estimate exposure in the near-road environment. A number of different dispersion models have been used in previous studies to characterize the impact of mobile source emissions. In addition to R-LINE, C-LINE and

AERMOD include the single source emissions based on simulated emissions inventories. Dispersion models have also been used in conjunction with chemical transport models to utilize both models strengths. Additional research should continue to understand the strengths of dispersion models as well as their weaknesses in order to improve the accuracy of the model results at different time scales and resolutions.

As this dissertation showed, the location of a monitoring site and the natural or build environment around the site drives the measured concentrations. To improve the accuracy of model estimations, these site characteristics need to be included and therefore well understood. The latest version of R-LINE allows the input of certain built environment features to be included in the model inputs. While these model features begin to address the bias observed in the model concentration estimates, other physical site or source characteristics should be considered for small domain modeling. R-LINE is also designed to accept a variety of meteorological inputs. The simulations presented within this dissertation should be compared to simulations using meteorological data collected at the pollutant monitoring site. This comparison would help show the sensitivity of the model results to the meteorology location used.

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